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CONTENTS.

F. ROELS and L. MOLL: "On the Index Loquelae". (Communicated by Prof. H. ZWAARDEMAKER), p. 98.

C. A. CROMMELIN, J. PALACIOS MARTINEZ and H. KAMERLINGH ONNES: "Isothermals of monatomic substances and their binary mixtures. XX. Isothermals of neon from + 20° C. to —217° C", p. 108.

A. SMITS: "Metals and Non-Metals". (Communicated by Prof. P. ZEEMAN), p. 119.

H. P. BARENDRECHT: "Urease and the radiation-theory of enzyme-action", p. 126.

P. N. VAN KAMPEN: "On the phylogeny of the hair of mammals". (Communicated by Prof. WEBER), p. 140.

G. NORDSTRÖM: "Note on the circumstance that an electric charge moving in accordance with quantum-conditions does not radiate". (Communicated by Prof. H. KAMERLINGH ONNES), p. 145.

L. E. J. BROUWER: "Remark on Multiple Integrals", p. 150.

Physiology. — *"On the Index Loquelae"*. By F. ROELS and L. MOLL. (Communicated by Prof. H. ZWAARDEMAKER).

(Communicated in the meeting of December 28, 1918).

The idea index vocalis has been introduced by GRADENIGO [1]) and denotes the quantitative relation between whispers of moderate energy and ordinary speech. Practically this relation is best determined with the aid of the extreme distances at which they are audible. According to GRADENIGO the average index for vowels is $^1/_7$ or $^1/_8$ [2]). According to WOLF [3]), ZWAARDEMAKER [4]), and REUTER [5]) the average index ranges from $^1/_5$ to $^1/_7$. For twenty monosyllabic, aequiintensive, isozonal test-words ZWAARDEMAKER—QUIX [6]) and REUTER [7]) found the index $^1/_{12}$.

The determination of the relative energy of whispers and ordinary speech being rather difficult (as it is difficult to distinguish exactly between what is meant by the one and what by the other), ZWAARDEMAKER has had recourse to RALEIGH's mirror for sound-measurement [8]). It then appeared that the normal average index vocalis $\frac{\text{reserve-air whisper}}{\text{ordinary speech}} = {}^1/_{16}$, the normal average index vocalis $\frac{\text{stage-whisper}}{\text{ordinary speech}} = {}^1/_4$. These indices enable us to reduce the whisper-values obtained for vowels and words to ordinary speech-values.

In experimenting on the intensity of audition separate words are used which consist of isozonal and aequi-intensive sounds, which, for

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[1]) GRADENIGO. Communication au congrès internat. d'otologie. Budapest 1909 GRADENIGO et STEFANINI. Sur l'acoumétrie. Propositions et études. Archives internat. de laryngolie, d'otologie et de rhinologie, 1911.

[2]) ZWAARDEMAKER. Over den index vocalis bij keuringen en bij de studie der ziektebeelden. (Handelingen van het XIVe. Ned. Nat.- en Gen. Congres te Delft 1913), p. 370.

[3]) WOLF. Sprache und Ohr. 1870. Frankfurt.

[4]) ZWAARDEMAKER Ibid.

[5]) REUTER. Zeitschr. f. Ohrenheilkunde. Bd. 47, blz. 91; Onderzoekingen Physiol. Lab. Utrecht (5) Vol. 5. p. 239.

[6]) ZWAARDEMAKER. Over geluidmeting. Zittingsverslag dezer Academie van 23 April 1915. Vol. XXIII. p. 1405.

[7]) REUTER. Onderzoekingen Physiol. Lab. Utrecht (5) Vol. 5. p. 249.

[8]) ZWAARDEMAKER and QUIX. De studie van het spraakgehoor. Onderzoekingen Physiol. Lab. Utrecht (5) Vol. 5. p. 1.

the very reason that every component of the word is heard as distinctly as the others, excludes guessing almost altogether and facilitates the finding of a definite zone of the scale. The question arises, however, though it is not probable, whether the hearing distance extends for whispered sentences as far as for words and vowels. ZWAARDEMAKER believes on the ground of various experiences that the distance at which whispered sentences are heard in a closed space amounts to 1 to 3 m.[1]) according to the words that are chosen. He calls the relation between this average distance and that at which whispered sounds are heard: *index loquelae.*

In the present investigation we purposed originally only to determine the index loquelae. However, the combination of the whisper-method with that of the systematic or experimental introspection, procured us a number of data of some importance for the psychology of perception. We availed ourselves of these data only in so far as they influence the determination of the index loquelae, the subject proper of this paper.

The material used by us consisted of rather simple sentences. The shortest consisted of 3, the longest of 14 syllables. Most often the sentences were in the indicative mood, seldom in the subjunctive mood or in the imperative mood. Though the present tense was used most frequently, we sometimes also employed the past and the future. Negative sentences were rare. In selecting our material we took an adequate number of symmetrical sentences, i.e. sentences having the same number of syllables before and after the copula, which consisted of one syllable, or an even number of syllables, or we chose such sentences as were divided by a word or by appropiate punctuation into two parts of an equal number of syllables (e.g. waste not, want not; a friend in need is a friend indeed). In most sentences there was a subject, a predicate and an object in the given order; sentences without one of these parts or with a verb split up as the construction required, we term irregular sentences. Proverbs and proverbial sayings were seldom used; sometimes, but not often, a French or Latin sentence was presented to the subject. All in all we made 328 experiments with 4 subjects (R., M., D. and A.). Table I gives a survey of the material with respect to the number of syllables, the symmetry and the asymmetry, the regular or the irregular construction of the sentences. It should be noted, that some test-sentences have

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1) ZWAARDEMAKER. Ueber die Anwendung von Sig. Exners Akustik von Hörsälen auf die Theorie der medizinischen Hörapparate. Wiener Medizinische Wochenschrift. Nr. 14, 1916.

TABLE I.

Subj.	Number of Ex.	3 s.	4 s.	5 s.	6 s.	7 s.	8 s.	9 s.	10 and more sentences.	Symm. sentences	Irregular sentences.	Proverbs.
R.	153	2	17	25	33	24	27	14	11	26	21	21
M.	74	2	12	12	14	14	7	2	11	12	23	12
D.	86	1	15	12	29	14	11	3	1	12	7	—
A.	15	—	—	—	—	2	5	3	5	4	4	2
Total.	328	5	44	49	76	54	50	22	28	54	55	35

TABLE II.

	3 s.			4 s.			5 s.			6 s.			7 s.			8 s.			9 s.			10 and more s.		
Subj.	M.	M.D.	C.V.	M.	M.D.	C.V.	M.	M.D.	C.V.	M.	M.D.	C.V.	M.	M.D.	C.V.	M.	M.D.	C.V.	G.	M.D.	C.V.	M.	M.D.	C.V.
R.	4	—	—	3.88	1.12	4	3.33	1.08	3	3.18	1.10	3	3.74	0.90	4	3.50	0.89	3	4.11	0.82	4	3	0.50	3
M.	5	—	—	3.71	0.91	4	3.12	1.06	3	2.64	0.73	2.5	2.71	0.91	3	2.35	0.71	2	2.25	—	—	1.83	0.67	1.5
D.	5	—	—	3.23	1.04	3	4.37	0.73	5	3.50	1.17	4	3.82	0.79	4	3.77	1.02	4	3.67	—	—		—	—

contributed values for two of the last three columns; a proverb like "a friend in need is a friend indeed" contributed to the making of the column of proverbs as well as to that of the symmetrical sentences.

The material was presented in whispers with reserve air. The subject sat ready to receive the whispers with the right ear, vertical to a line from this ear to the mouth of the experimenter. The maximal distance was 5 m.; if the subject failed to correctly repeat the spoken words at that distance, it was lessened every time by 1 m. until the spoken words were reproduced correctly. When the distance was reduced to 2 m., it was lessened every time by $^1/_2$ m. Careful introspection, at which the subject recorded minutely what he had heard, followed after each stage of the experiment. At every sitting 12 experiments were made at most, to prevent fatigue. They were all carried out in the over-furnished library of the Utrecht Clinic for Psychiatry and Neurology. Before starting the experiments proper, we determined the audition of our subjects by means of the words given by ZWAARDEMAKER and QUIX in "De Studie van het Spraakgehoor" [1]). It proved to be normal with all of them; the hearing-distance varied for the words employed from 6 to 30 m.

In Table II we have calculated the mean distances (M.), for every one of the three snbjects R. M. and D., at which sentences of the given number of syllables were heard (the values procured by A. are left out of account as being too small in number).

As precision-indices we also give the mean deviations (M. D.) and the central values (C. V.).

It is obvious that with a larger number of syllables the sentences are heard less distinctly. For R. M. and D. this difference is resp. 1, 3.17 and 1.33 m., in the case of sentences of 3 to 10 or more syllables. Besides to the influence of factors which we cannot discuss further here, these considerable individual differences are to be ascribed chiefly to the various tasks imposed upon the subjects. It stands to reason that proverbs and proverbial sayings are generally heard at greater distances than non-proverbial expressions; symmetrical sentences farther than asymmetrical sentences; on the other hand irregular sentences are more difficult to hear than those that are constructed regularly. Table III (in which the frequency of the various kinds of sentences is tabulated in percentages of the total number of test-sentences presented) shows clearly that the intensity of audition varied considerably.

The difference of the distances at which sentences of 3 and those

[1]) Onderzoekingen Physiol. Lab. Utrecht (5) vol. 5 p. 1.

of 10 or more syllables are heard, is greatest with the subject M. (3.17 m.). This is owing to a considerable extent, to the high per-

TABLE III.

Subj.	Symmetrical sentences.	Proverbs.	Irregular sentences.
R.	17	13.7	13.7
M.	16.2	16.2	31.1
D.	14	—	8.1

centage of irregular sentences (31.3) with their unfavourable influence upon auditory sensation. This influence was much less on R. and D. (resp. 13.7 and 8.1 %). The comparatively small difference in the case of R. (1 m.) is to be ascribed to the circumstance that symmetrical sentences and proverbs constitute about $^1/_3$ (30.7 %) of the total number of the test-sentences that were used. D., being a FLEMING and consequently unacquainted with Dutch proverbs (which were not presented to him) is placed between R. and M., quite in accordance with the difference in the hearing distance with 14 % favourable and 8.1 % unfavourable factors.

When we look at Table II more closely, it appears that intra-individual differences are not less conspicuous than the inter-individual ones. For, though it is true that the hearing distance with the three subjects for trisyllabic sentences is considerably greater than for sentences of 10 or more syllables, the distance decreases far from regularly. In the case of M. the decrease is gradual with only few exceptions.

Sentences of an odd number of syllables are generally heard better by R. and D. than those of an even number having one syllable more, they even perceive sentences of 9 syllables resp. 0.23 m. and 0.44 m. farther than those of 4 syllables.

The cause of this phenomenon, so strange at first, is the fact that a symmetrical construction, which largely promotes our hearing faculty, occurs more often with sentences of an odd number of syllables than with those of an even number. This is because with all symmetrical sentences the number of syllables before and after the copula is the same, while with most of them the copula is made up of one syllable (is, was, had, are etc.) Besides the agreement just alluded to there is also some difference in the shifting of the hearing istance, when the number of syllables increases. In the case of R.

the distance generally increases with an increase of the number of syllables from 4 to 9, for sentences of an odd number as well as an even number — with a few exceptions in the latter case. With D., however, a diminution is observed with sentences of an odd number of syllables, an increase with those of an even number. We have seen, then, that while in the case of M the distance decreases regularly (with a few exceptions) from 4 to 9 syllables, with R. it increases for an odd as well as for an even number of syllables; with D. an increase is observed for the odd and the decrease for the even numbers.

What is the cause of the increase of the hearing distance with an increase of the number of syllables in the cases of the subjects *R* and *D*, especially the first; and what gives rise to the differences in the subjects? One might expect that with the increase of the number of syllables the retro-active inhibition is more keenly felt, in consequence of which the faculty of perception is weaker.

Audition, however, results from the joint action of apperception and assimilation; the latter, just as in the case of reading, plays a prominent part here. Now the process of assimilation requires a minimum of apperceived elements; it can readily be imagined that in sentences of a small number of syllables such elements are as a rule too small in number to serve as starting-points for the assimilation-process. The assertion that with shorter sentences the assimilation-process should naturally be less extensive and should therefore require fewer points of contact, does not hold good. We have observed again and again that a certain number of apperceived elements, no matter how short the sentences may be, is required to prevent assimilation from becoming mere guessing. So the number of apperceived elements required for test-sentences of various length is not proportionate to the increase of the number of syllables. It follows that the reproduction-tendencies emanating from the apperceived elements are, within certain limits, stronger than the inhibition they exert upon each other. The reverse seems to be the case with all subjects for sentences of ten or more syllables.

It is now easy to explain also the individual differences. With M., for whom we observe a gradual decrease of the hearing distance with an increase of the number of syllables, the influence of inhibition is stronger than that of assimilation. With R., for whom the hearing distance increases with the length of the sentences of an odd as well as an even number of syllables, the reverse obtains, viz. the unfavourable effect of inhibition is not only arrested, but with the increase of the number of points of contact, assimilation

also acts more intensely. Subject D. takes up a place between R. and M. in that for sentences of an even number of syllables assimilation gets the start of inhibition when the sentences get longer, whereas for sentences of an odd number of syllables inhibition proves the stronger. We are unable to account for this strange phenomenon.

By means of the normal average index vocalis $\frac{\text{reserve-air whisper}}{\text{ordinary speech}} = {}^{1}/_{18}$, which index presumably also holds for whispered sentences, we have reduced in Table IV the whisper-values for sentences of various length to ordinary-speech values. Both sorts of values in Table IV constitute the average of the hearing distances, determined for the subjects R., M., and D., with respect to the length of the various sentences.

TABEL IV.

Syllables.	Whispers.	Loud speech.
3	4.67	60.71
4	3.61	46.93
5	3.61	46.93
6	3.11	40.43
7	3.42	44.46
8	3.21	41.73
9	3.34	43.42
10 and more	2.41	31.33

Be it observed by the way that owing to the more conspicuous survey of the data in Table IV, our conclusions deduced from Table II are shown here to a greater advantage viz, the great difference between the hearing distances for sentences of 3 and for those of 10 or more syllables (loud speech nearly 30 m.); the comparatively slight difference for sentences of 4 to 9 syllables (loud speech ± 3.5 m); the greater distance for sentences of an odd number of syllables, owing to their most often symmetrical construction; etc.

We have already pointed out that symmetrical sentences are heard at greater distance than asymmetrical sentences. Table V shows that symmetry is highly favourable to our audition. Here also great individual differences are observed; for R., M. and D. they are resp. 0,61; 1.13; and 0.24 m.

It is easy to understand that proverbs and proverbial sayings are heard at greater distance. The subjects' familiarity with them, helps

TABLE V.

	Symmetrical sentences			Asymmetrical sentences
Subj.	M.	M.D.	C.V.	M.
R.	4.11	0.79	4	3.50
M.	3.87	1	4	2.57
D.	4.21	0.96	4.5	3.97

the assimilation process consequent on the apperception of elements.

The favourable influence exerted by proverbs and proverbial sayings is, however, inferior to that of symmetrical construction, as is borne out by a comparative study of Tables V and VI. The enlargement of the hearing distance for symmetrical sentences is with R. and M. respectively 0.61 and 1.13 m., that of hearing proverbs only 0.74 m.

TABLE VI.

	Proverbs.			The other sentences.
Subj.	Mean.	M.D.	C.V.	M.
R.	3.78	1	4	3.37
M.	3.67	0.94	4	2.93

From a study of the data of Table VII it also appears that the absence of subject, predicate or object or an inversion of these parts of a sentence may be an impediment to hearing (see page 106). Irregular sentences are heard by R., M. and D. at a much shorter distance than the regular ones. The decrease of distance is resp. 0.73; 0.28; and 1.01 m. The very considerable decrease with D. is probably owing to the circumstance that as a Fleming he is less familiar with the rather uncommon construction of many of our irregular sentences.

The relation between the mean distances at which whispered sentences and whispered words are heard we term after ZWAARDEMAKER index loquelae. In otiatrics the distance at which the latter are heard is estimated at ± 18 m.

In their experiments ZWAARDEMAKER and QUIX [1]) arrived at the same conclusion.

TABLE VII.

	Irreg. sentences.			Reg. sentences.
Subj.	M.	M.D.	C.V.	M.
R.	2.90	0.72	3	3.63
M.	2.65	1.11	2.75	2.93
D.	2.64	0.88	2.50	3.65

The mean distance at which whispered sentences of from 3—14 syllables are heard amounts to ± 3.42 m, as shown in Table II and IV, so that the index loquelae is $\frac{3.42}{18} = {}^{19}/_{100}$.

SUMMARY.

1. Our material consisted of sentences of 3 to 14 syllables. They were presented by means of whispers with reserve-air.

2. We determined the intensity of hearing of our subjects by means of separate words, composed of isozonal and aequi-intensive sounds. With all of them audition appeared to be normal; the distances for different words varied from 6 to 30 m.

3. The difficulty of hearing sentences increases with the number of syllables. The difference in the distances at which sentences of 3 syllables and those of 10 or more syllables are heard, averages 1.83 m. (whispers).

4. Generally the decrease of the distance in proportion to the increase of the number of syllables does not take place gradually. Sentences of an odd number of syllables are as a rule heard better than those of an even number containing one syllable more. Even sentences of 9 syllables are heard on an average 0.33 m. farther than those of 4 syllables. The reason is that symmetrical construction, which aids our hearing faculty, occurred more with an odd than with an even number of syllables.

5. Proverbs and proverbial sayings are heard at larger distances. However, the favourable influence of proverbs and proverbial sayings

[1]) De studie van het spraakgehoor. Onderzoekingen Physiol. Lab. Utrecht (5) dl. 5 blz. 1.

is inferior to that of mere symmetrical construction of the sentences.

6. The absence of subject, predicate or object or inversion of the order of words are an impediment to audition; the hearing distance is lessened by it.

7. Hearing is the result of the joint action of perception and assimilation. For sentences of 3 to 9 syllables the influence of the reproduction-tendencies, consequent on the apperceived elements, counterbalances that of the inhibition, caused by an increase of the number of syllables. Only for sentences of 10 and more syllables the latter seems to get the start of the former.

8. The index loquelae i.e. the relation between the mean distances at which whispered sentences and whispered words are heard is $^{19}/_{100}$.

Physics. — "*Isothermals of monatomic substances and their binary mixtures. XX. Isothermals of neon from + 20° C. to —217° C.*" By C. A. CROMMELIN, J. PALACIOS MARTINEZ, and H. KAMERLINGH ONNES. Communication N°. 154*a* from the Physical Laboratory at Leiden.

(Communicated in the meeting of June 29, 1918).

§ 1. *Introduction.* This paper is the continuation of a previous preliminary communication [1]). The reduction of the neon-isothermals has now progressed so far, that what follows may be looked upon as a pretty nearly completed whole. The measurements refer to pressures up to about 90 atmospheres and as regards the temperature go from + 20° C. down to — 217° C.; they therefore embrace the region between the ordinary temperature down to the lowest temperature to be reached with liquid oxygen. The region from — 218° C. to —246° C., which can now also be covered by means of the hydrogen vapour cryostat [2]), is here left out of account; we hope shortly to be able to continue our measurements in this region. As regards the importance of such determinations as will be communicated on this occasion and the apparatus which have been used for the purpose we may refer to previous communications on the isothermals of hydrogen and of argon. [3])

For the sake of completeness and of a better survey of the whole work we have included in the table the material published in the previous communications on neon, quoted above, viz. the series I, II, III, IV, VI, VII, VIII and IX.

§ 2. *The results of the measurements* are given in table I, where

$\theta =$ the temperature on the *international* [4]) CELSIUS-scale, i.e. the temperature on the *international* KELVIN-scale diminished by 273.09;

$p =$ the pressure in international atmospheres (for Leiden 1 international atmosphere = 75.9488 cms mercury);

$d_A =$ the density expressed in the normal density (0° C. and 1 atm.);

$v_A =$ the volume expressed in the normal volume (0° C. and 1 atm.).

[1]) H. KAMERLINGH ONNES and C. A. CROMMELIN, Comm. N°. 147*d*; these Proc. XVIII (1) p. 515.

[2]) H. KAMERLINGH ONNES, Comm. N°. 151*a*; these Proc. XIX (2) p. 1049.

[3]) Comp. H. KAMERLINGH ONNES and H. H. FRANCIS HYNDMAN, Comm. N°. 69; these Proc III p. 481; H. KAMERLINGH ONNES and C. BRAAK, Comm. N°. 97*a* these Proc. IX p. 754; C. BRAAK, Dissertation, Leiden, 1908; C. A. CROMMELIN, Dissertation, Leiden, 1910.

[4]) Comp. H. KAMERLINGH ONNES, Comm. Suppl. N°. 34*a*. § 5.

TABLE I.

Series.	N°.	p	d_A	pv_A
		$\theta = +20°.00$ C.		
VI	1	22.804	21.046	1.0835
VI	2	25.015	23.052	852
VI	3	26.575	24.464	863
VI	4	29.090	26.757	872
VI	5	32.572	29.891	897
VIII	1	34.887	32.002	902
VI	6	35.423	32.447	917
VI	7	37.812	34.601	928
VIII	2	39.168	35.843	928
VIII	3	44.762	40.862	955
VIII	5	54.149	49.213	1003
VIII	6	59.717	54.161	026
VIII	7	65.021	58.797	059
VIII	9	77.360	69.338	131
VIII	10	82.545	73.967	160
VIII	11	88.239	78.886	186
VIII	12	93.298	83.154	220
		$\theta = 0°.00$ C.		
VII	1	22.064	21.869	1.0089
VII	2	23.555	23.314	103
VII	3	25.867	25.558	121
VII	4	28.468	28.089	135
VII	5	30.790	30.345	147
IX	1	39.753	39.098	168
IX	2	44.892	44.030	196
IX	5	59.777	58.234	265
IX	6	66.104	64.135	307
IX	7	74.059	71.495	359
IX	8	79.108	76.127	392
IX	9	84.662	81.347	408

TABLE I (*Continued*).

Series.	N°.	p	d_A	pv_A
		$\theta = -103°.01$ C.		
XV	1	35.558	56.40	0.6304
XV″	1	36.697	58.23	6302
XV	2	40.610	64.21	6324
XV″	2	42.107	66.53	6329
XV″	4	55.136	86.57	6369
XV	4	58.583	91.76	6384
XV	5	78.110	120.52	6481
		$\theta = -141°.22$ C.		
XVI″	1′	33.840	69.83	0.4846
XVI	2	37.707	77.71	4852
XVI″	2	38.581	79.50	4853
XVI	3	43.319	88.97	4869
XVI″	4	49.881	102.32	4875
XVI	4	51.916	106.42	4878
XVI	5	66.471	134.91	4927
XVI	6	78.558	158.06	4970
		$\theta = -182°.60$ C.		
X	2	32.067	99.89	0.3210
X″	2	32.988	102.84	3208
X	3	36.438	113.69	3205
X″	3	36.880	115.07	3205
X	4	41.371	129.44	3196
X″	4	42.533	133.15	3194
X	5	49.943	156.61	3189
X″	5	50.514	158.55	3186
X″	6	63.320	199.21	3179

TABLE I (*Continued*).

Series.	N°.	p	d_A	pv_A
		$\theta = -200°.08$ C.		
XI	1′	26.214	105.10	0.2494
XI	2′	28.402	114.38	2483
XI	3″	31.417	127.24	2469
XI	1	34.268	139.81	2451
XI	4	34.285	139.88	2451
XI	5	39.843	164.30	2425
XI	2	39.891	164.63	2423
XI	3	46.517	194.30	2394
XI	3′	46.529	194.51	2392
XI	6	47.951	200.79	2388
III	1	61.657	263.77	2338
III	2	67.456	291.10	2317
III	3	73.850	320.35	2302
III	4	79.923	348.59	2293
		$\theta = -208°.10$ C.		
XII	1	24.071	111.90	0.2151
XII	3	28.844	136.44	2114
XII	4	31.948	153.00	2088
XII	5	37.856	185.47	2041
XII	6	41.798	207.95	2010
IV	1	58.472	308.32	1897
IV	2	64.451	345.22	1867
IV	3	69.692	377.89	1844
IV	4	74.532	409.18	1822
IV	5	79.228	439.12	1804

TABLE I (*Continued*).

Series.	N°.	p	d_A	pv_A
		$\theta = -213°.08$ C.		
XIII	1	23.086	119.92	0.1925
XIII	2	24.810	129.82	1911
XIII	3	26.673	140.90	1893
XIII	4	29.365	157.70	1862
XIII	5	32.441	177.37	1829
XIII	6	37.418	210.68	1776
II	1	53.896	334.59	1611
II	2	59.769	382.03	1565
II	3	66.271	435.46	1522
II	4	72.858	484.75	1503
II	5	79.698	534.62	1491
		$\theta = -217°.52$ C.		
XIV	1	21.349	123.40	0.1730
XIV	2	22.997	134.72	1707
XIV	3	24.686	146.67	1683
XIV	4	26.848	162.51	1652
XIV	5	30.042	186.94	1607
XIV	6	32.795	209.68	1564
I	1	49.930	358.51	1393
I	2	53.528	395.62	1353
I	3	59.618	458.40	1301
I	4	64.975	511.85	1269
I	5	71.649	571.69	1253
I	6	79.417	632.23	1256

A graphical representation of the observations will be found in fig. 1 in a $\left(\frac{pv_A}{T}, d_A\right)$ diagram.

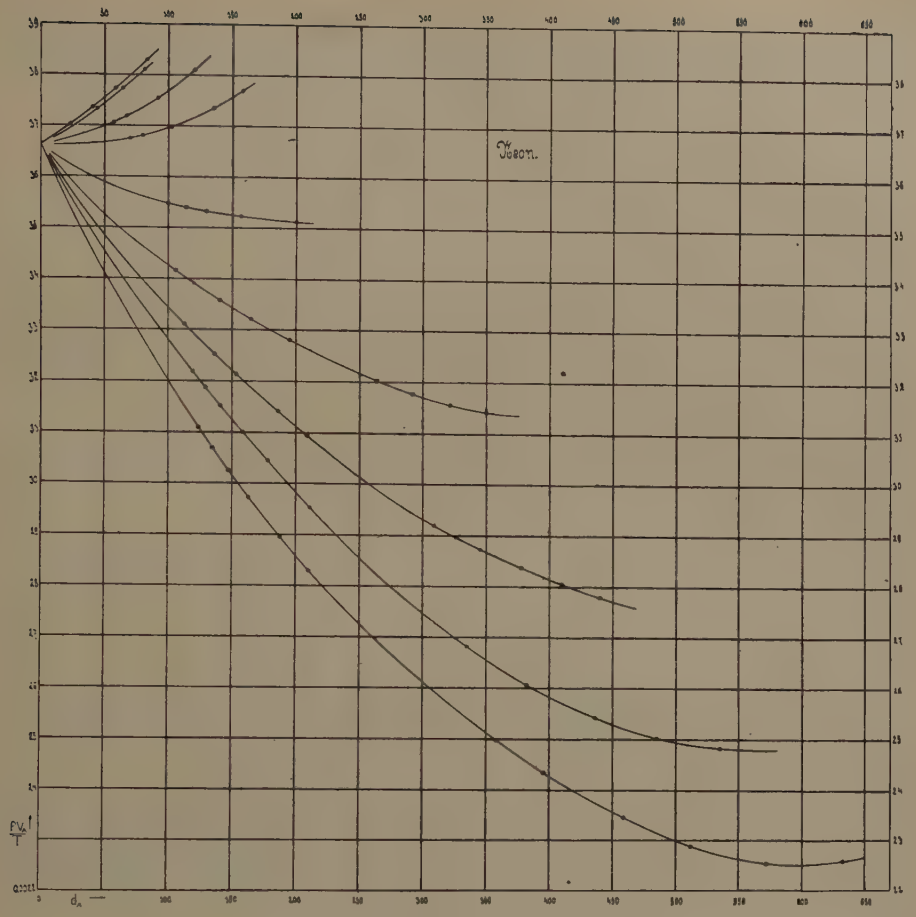

Fig. 1.

§ 3. *Virialcoefficients.*

By means of the above results some of the coefficients in the empirical equation of state

$$pv_A = A_A + B_A d_A + C_A d_A^2 + D_A d_A^4 + E_A d_A^6 + F_A d_A^8 \quad [1])$$

could be computed. These calculations only embrace the coefficients B_A, C_A, D_A and in one case E_A; for the densities which were reached are not so high as would be necessary for the deduction of F_A and in most cases of E_A also; these were therefore found from the reduced equation of state VII. A. 3 [2]) or VII. 1. [3]), in which the coefficients $\mathfrak{E}$ and $\mathfrak{F}$ are identical. In some of the calculations,

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[1]) H. KAMERLINGH ONNES, Comm. N°. 71; these Proc. IV p. 125.

[2]) H. KAMERLINGH ONNES and C. A. CROMMELIN, Comm. N°. 128; these Proc. XV (1) p. 273.

[3]) Suppl. N°. 19.

as will appear presently, D_A, in others D_A and C_A were assumed according to VII. A. 3. For this purpose use was made of the critical constants of neon as published on a former occasion [1])

$$\theta_k = -228°.35\ C. \qquad p_k = 26.86\ \textit{int. atm.}$$

The calculations were conducted in three different ways:

a. for all temperatures only B_A was calculated from the observations, the remaining coefficients being assumed, viz. C_A and D_A according to VII. A. 3., E_A and F_A according to VII. 1 or to VII. A. 3- (as noticed above, this comes to the same);

b. for the lowest 4 temperatures B_A and C_A were computed from the observations, further as under *a*;

c. for all temperatures B_A and C_A were deduced from the observations, for —200°.08 C., —208°.10 C. and —213°.08 C. also D_A and for —217°.52 C. also E_A, further as under *a*.

The calculations *c* were made first, with a view to obtaining the best possible accordance with the observations, the coefficients therefore bearing a purely empirical character. When it appeared that the values of C_A could not be connected by a smooth curve, much less those of D_A, which proved the observational material to be insufficient for the deduction of C_A and D_A as functions of the temperature, we proceeded to the methods given under *a* and *b*, in which the values of D_A and partly even those of C_A were assumed. Naturally the accordance with the observations is very much inferior with the methods *a* and *b* than with *c*.

The results of the calculations which were all conducted by the method of least squares are found in tables II and III. Table II gives the individual virial-coefficients, as calculated from the observations according to *a*, *b*, and *c*, table III the coefficients borrowed from VII. A. 3 as well as the values of A_A computed from the equation

$$A_A = A_{A_0}(1 + 0.0036618\,\theta),\ ^{2})$$

where for A_{A_0} the value + 0.99986, as published on a former occasion [3]), was taken as a basis.

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[1]) H. Kamerlingh Onnes, C. A. Crommelin and P. G. Cath, Comm. N°. 151*b*, these Proc. XIX (2) p. 1058.

[2]) Comm. N°. 71.

[3]) Comm. N°. 147*d*.

TABLE II.

θ	$B_A \times 10^3$	$B_A \times 10^3$	$C_A \times 10^6$
	According to *a*.	According to *b*.	
+ 20°.00	+ 0.54880		
0°.00	47148		
— 103°.01	16653		
— 141°.22	055249		
— 182°.60	— 0.093113		
— 200°.08	15746	— 0.18779	+ 0.21531
— 208°.10	19553	21706	18307
— 213°.08	22305	24084	18407
— 217°.52	24028	25880	19649

TABLE II (*Continued*).

θ	$B_A \times 10^3$	$C_A \times 10^6$	$D_A \times 10^{12}$	$E_A \times 10^{18}$
	According to *c*.			
+ 20°.00	+ 0.51578	+ 0.82778		
0°.00	41334	1.1538		
— 103°.01	069193	1.1515		
— 141°.22	— 0.025378	0.71945		
— 182°.60	13435	33607		
— 200°.08	19667	27847	— 0.24096	
— 208°.10	22926	25304	0.16102	
— 213°.08	24625	21123	0.005848	
— 217°.52	29313	36427	0.46739	+ 0.57517

TABLE III.

θ	A_A	$C_A \times 10^6$	$D_A \times 10^{12}$	$E_A \times 10^{18}$	$F_A \times 10^{27}$
		According to VII. A. 3.			
+ 20°.00	+ 1.0731	+ 0.29747			
0.00	0.99986	25440			
— 103°.01	62271	0.072156	+0.37445	— 0.1373	+ 40.29
— 141°.22	48281	39576	28409	0.03754	12.51
— 182°.60	33131	58524	12718	2409	— 4.190
— 200°.08	26731	96581	11124	4293	5.666
— 208°.10	23795	0.12219	0.081145	+0.04550	5.367
— 213°.08	21971	14073	60843	4599	4.836
— 217°.52	20345	15882	41215	4576	4.160

§ 4. *Discussion and comparison with other observations.* The differences between the pv_A-values calculated from these equations and the observed values are represented graphically in fig. 2 as functions of the densities d_A, the ordinates being the observed minus the calculated pv_A-values, expressed as percentages of the latter. In this manner the character of the deviations is more easily grasped than would be the case, if the numbers were given in the tables.

The correspondence between the new and the old series is very satisfactory on the whole; only in the isothermal for — 217°.52 C. a marked deviation may be noticed. Whereas for the isothermals of —200°.08 C. the deviations of the observations from the most closely corresponding formula (method *c*) are within 0.1 %, differences of almost $^1/_2$ % occur in the isothermals for — 217°.52 C.

The differences between the various sets of B_A-values obtained on this occasion from the smoothed B_A-values according to VII. A. 3, viz. $\Delta B_A = B_A$ (calc.) $- B_A$ (VII. A. 3) are represented in fig. 3; the corresponding deviations of the B_A-values obtained by CATH and one of us[1]) from measurements at low pressures are included in the figure.

[1]) P. G. CATH and H. KAMERLINGH ONNES, Comm. N°. 152*e*, presented to the Meeting of the Academy some time ago and shortly to be published in the Proceedings; preliminary values are given by P. G. CATH, Dissertation, Leiden 1917, p. 77.

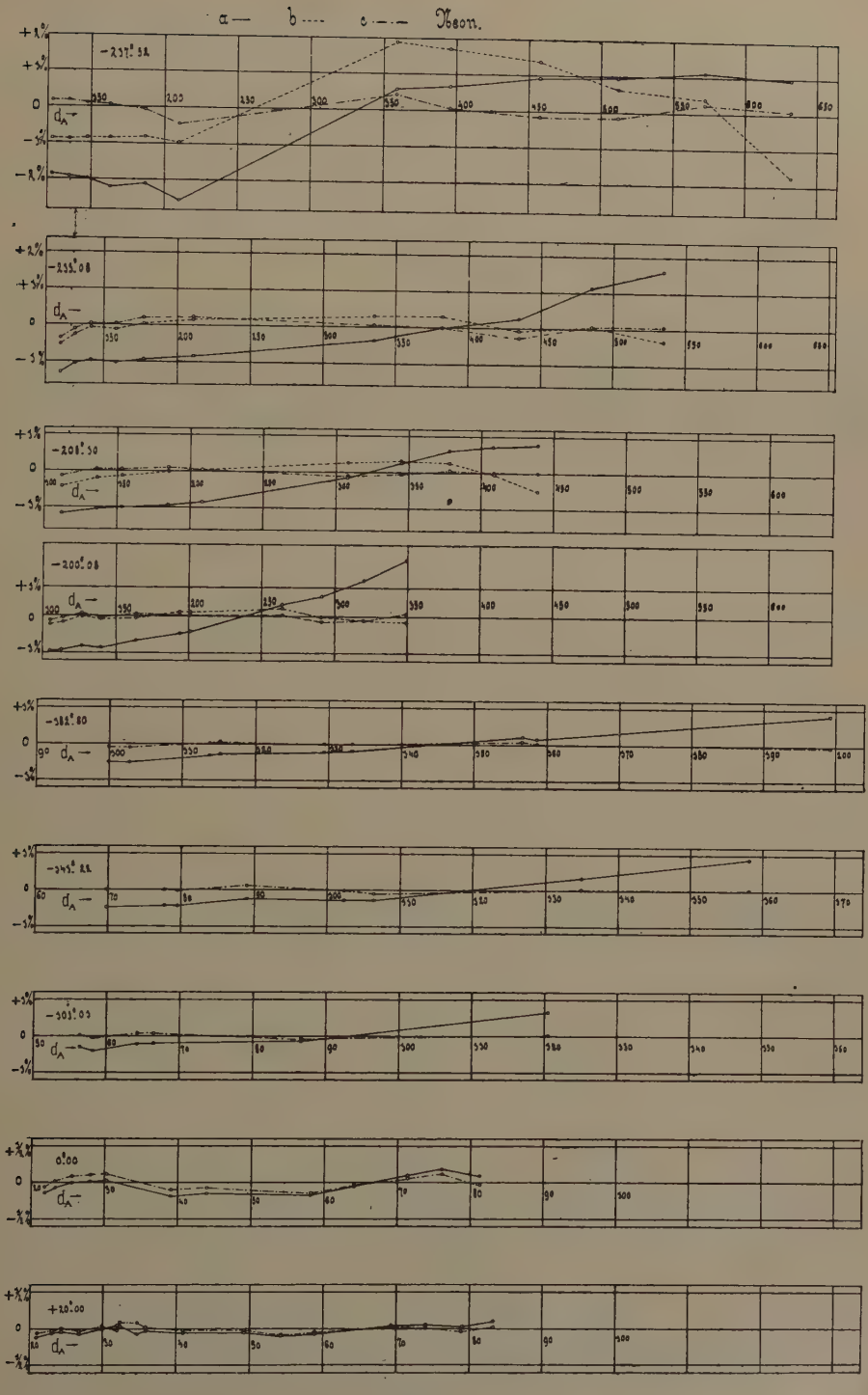
a— b---- c-.-- Neon.
-237°.52
-233°.08
-208°.50
-200°.08
-382°.80
-349°.82
-303°.03
0°.00
+20°.00
d_A

Fig. 2

It may be noticed, that the B_A-values according to a agree fairly

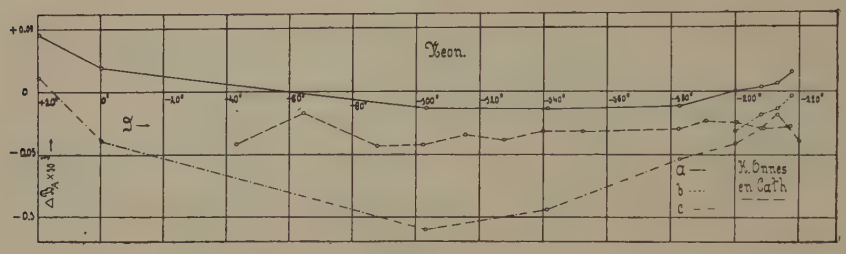

Fig. 3.

well with those according to VII. A. 3. Those obtained by method c deviate much more markedly, as might be expected; especially at the lower temperatures they show much smaller values.

Chemistry. — "*Metals and Non-Metals*". By Prof. A. SMITS. (Communicated by Prof. P. ZEEMAN).

(Communicated in the meeting of May 3, 1919).

Introduction.

In a few previous communications I already discussed the unattackable electrodes and their efficiency as gas-electrodes. Only the hydrogen electrode was, however, discussed in detail. It was pointed out that the unattackable electrodes are among the most inert metals, being so inert that even in contact with an electrolyte they do not assume internal equilibrium, so that they are almost always in disturbed condition, and we do not even know the potential of the really unary metal.

When such a metal is immersed in the aqueous solution of an acid, while hydrogen is passed through, the electron concentration of the metal equilibrium in the electrolyte, which we represent by the equation:

$$M_L \rightleftarrows M_L^{\nu\cdot} + \nu\theta_L$$

becomes equal to the electron concentration of the hydrogen equilibrium:

$$H_{2L} \rightleftarrows 2H_L^{\cdot} + 2\theta_L$$

in agreement with the pressure of the hydrogen that is passed through, which means that the hydrogen phase and the hydrogen-containing metal-phase possess the same potential, the electromotive force of the circuit metal-electrolyte-hydrogen being given by the equation:

$$E = -\frac{RT}{F} ln \frac{(\theta_L)_M}{(\theta_L)_{H_2}} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

Hence the hydrogen-electrode indicates the potential belonging to the three-phase equilibrium metal phase + hydrogen phase + electrolyte in accordance with the prevailing pressure, temperature, and total ion concentration $(H^{\cdot}) + (M^{\nu\cdot})$. As, however, as was already stated in the discussion of the E,X-fig., the said electrolyte contains an entirely negligible concentration of ions of the so-called unattackable electrode, we may substitute the words: hydrogen-ion-concentration for "total-ion-concentration"

When we now consider the case that an unattackable electrode is placed in an electrolyte, chlorine being led through, we get the same thing in so far that the electron concentration of the metal equilibrium in the electrolyte is entirely dominated by the electron concentration of the chlorine equilibrium:

$$Cl_2 + 2\theta \rightleftarrows 2Cl' \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

in agreement with the pressure of the chlorine that is led through, from which follows that the chlorine-gas phase and the chlorine-containing metal phase will possess the same potential with respect to the electrolyte. Our more recent views about the electromotive equilibria lead to the assumption of an electro-ionisation equilibrium in chlorine gas, though the concentration of ions and electrons in this gas-phase is exceedingly small. That there exists such an electro-ionisation for chlorine, is proved by the exceedingly slight electrical conductivity. Now, however, the above mentioned equilibrium (2) does not suffice, for in electrically neutral chlorine this equilibrium cannot occur *alone*, because here there is only question of particles charged negatively electrically. Besides the chlorine electrode could not assume a positive charge with respect to the electrolyte.

Undoubtedly this difficulty has also been felt in the former view about the electromotive equilibrium. The negative charge which zinc assumes on immersion into an electrolyte was explained as follows: The zinc tends to go into solution as zinc-ion, and it has evidently always been imagined that this happened through a *simultaneous* splitting up of the zinc-atom into zinc-ion + electrons, in which, however, the zinc-ions only went into solution, and the negatively charged electrons remained on the metal.

When we wish to account for the positive charge which the chlorine electrode assumes with respect to an electrolyte in an analogous way, we get into serious difficulties, which have led to the assumption by some physicists that besides the free negatively charged electrons there exist also free positively charged electrons, and that these could render important services for the chlorine electrode and for the other non-metal electrodes. For just as the splitting up

$$Zn \rightarrow Zn^{\cdot\cdot} + 2\theta$$

was assumed when the zinc went into solution, the process

$$Cl_2 \rightarrow 2Cl' + 2\oplus$$

was supposed to take place at the chlorine electrode when chlorine went into solution. The chlorine ions formed go into solution and the positive electrons would be left behind in the chlorine electrode

This solution should, however, be denied all signification, because as yet free positive ions have not been met with, and everything points to the existence of only one kind of electrons, viz. the negative ones.

Hence in spite of a desperate attempt, the difficulty remained undiminished of force.

The Chlorine-Electrode.

When considering the just-discussed difficulty, I have come to the conclusion that we must necessarily assume that the chlorine atom possesses the power to split off electrons and to absorb them, and that these two processes take place side by side, which we can represent by the following equations: [1])

$$XCl_{2G} \rightleftarrows 2X\,Cl'_G + 2X\,\theta_G \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

and

$$YCl_{2G} + 2Y\theta_G \rightleftarrows 2YCl'_G \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

in which X and Y indicate the fractions of the original number Cl_2 mol. which have undergone a positive resp. negative ionisation.

As the electrons, which are absorbed according to (2) proceed from the electron-ionisation (1) it is clear that

$$Y \leq X.$$

In the limiting case $Y = X$ the chlorine would contain an equal number of positive and negative ions, and no electrons at all. As we have to do here with a non-metal, X and Y will be exceedingly small.

It is now the question how the positive charge of the chlorine electrode is to be explained. It is clear that for this we should have to assume that for the non-metal, chlorine, it is the negative ions that go practically exclusively into solution, and possess, therefore, a much greater solubility than the positive ones.

Metals and Non-Metals.

These considerations about the non-metal, chlorine, which necessarily result from the consistent application of the principles of the theory of allotropy to the electromotive equilibria, lead us to the point of view from which we can survey the metals and the non-metals in a very satisfactory way.

[1]) For the sake of simplicity we assume here that the positive ions are also univalent.

It is, indeed, clear that as the metallic and non-metallic properties in the periodic system of the elements gradually pass into each other, theory will have to make clear that between the metallic and the non-metallic state there exist only quantitative differences, and that there are therefore all kinds of intermediary states possible.

This demand can really be satisfied by an in every way plausible extension.

For this purpose we must assume that the atoms of all the elements, hence both metals and non-metals, can split off and absorb electrons so that the following reactions take place side by side:

$$XE_n \rightleftarrows X\frac{n}{m_1}E_{m_1}^{\nu_1} + X\frac{n}{m_1}\nu_1\theta \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

and

$$YE_n + Y\frac{n}{m_2}\nu_2\theta \rightleftarrows Y\frac{n}{m_2}E_{m_2}^{\nu_2'} \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

in which E_n denotes the molecule of an element and

$$E_{m_1}^{\nu_1} \text{ and } E_{m_2}^{\nu_2'} \text{ the ions.}$$

As is known for metals m_1 is mostly $= 1$, and probably m_2 will as a rule also be $= 1$. For non-metals m has often been found $= 1$, but several times also greater than 1. Of the factor m_1 no doubt the same thing may be expected.

With perfect certainty we may only say this that

$$Y\frac{n}{m_2}\nu_2 \leqq X\frac{n}{m_1}\nu_1 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

from which follows that when

$$\nu_1 = \nu_2$$

and

$$\nu_1 = \nu_2$$

the number of positive ions will be greater than the number of negative ones.

When now the question is put in what respects metals and non-metals will differ, the answer is as follows:

In the first place the factor X is comparatively great for metals and exceedingly small for non-metals, so that for non-metals also the factor Y is exceedingly small.

This is among other things in accordance with the great electric conductivity of metals and the exceedingly slight conductivity of non-metals.

In the second place for the metals the positive ions possess the greatest solubility, and for the non-metals the negative ions. This accounts among other things for the difference in electromotive

behaviour between metals and non-metals. In this it is noteworthy that the difference in solubility between the positive and the negative ions for the elements with exclusively metallic resp. exclusively non-metallic properties, is so great as to justify us in taking only positive resp. negative ions into account in the coexisting solution.

For the intermediate amphibious elements as Iodine, Sulphur, Selenium, Tellurium, Arsenic, and Antimony the existence of positive and negative ions also in solution, should certainly be taken into account to obtain a deeper insight, and here lies still an extensive field to be studied more closely, led by the more recent views about the electromotive equilibria.

Polarisation for non-metals.

When we indicate the non-metal by N, we may represent the formation of negative ions by the equation:

$$N_n + n\nu\theta \rightleftarrows nN^{\nu'} \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

Applying the well-known thermodynamic derivation for the potential difference, we get:

$$\Delta = \frac{RT}{\nu F} ln \frac{K'_{N^{\nu'}} N_P^{\nu'}}{(N_L^{\nu'})} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

in which $(N_P^{\nu'})$ represents the concentration of the negative ions in the phase of which the electrode consists, this may be either a gas, or a liquid, or a solid phase. Starting from the electron formula:

$$\Delta = \frac{RT}{F} ln \frac{K'_\theta (\theta_P)}{(\theta_L)} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

which is general, because the electron is the common constituent part of all matter, we may substitute for (θ_L) the value that follows from (6) for the liquid phase, applying the law of the chemical mass action; thus we arrive at the already known equation:

$$\Delta = \frac{RT}{n\nu F} ln \frac{K' (N_{n_L})}{(N_L^{\nu'})^n} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (9)$$

In order to study the polarisation phenomenon we cannot make use of equation (9), but we can use the newer equation (7). To get a good insight into this question it is desirable that we indicate the equilibria in the electrode completely; accordingly we must also express the splitting off of electrons, and write therefore:

$$X N_{n_P} \rightleftarrows X n N_P^{\nu'} + X n \nu \theta_P \quad . \quad . \quad . \quad . \quad . \quad (6a)$$

and

$$YN_{n_P} + Ynr\theta_P \rightleftarrows YnN^{\nu'}_P \quad . \quad . \quad . \quad . \quad . \quad . \quad (6b)$$

When we now make the non-metal electrode N anode, we withdraw the electrons. If we had to deal with a metal, hence if N was a metal, the electrode could maintain its potential in consequence of the fact that the withdrawn electrons are supplied in time by the reaction (6*a*), the positive ions formed going into solution. As N is however a non-metal, and the positive ions $N^{\nu'}$ do not practically go into solution, the reaction (6*a*) will not take place to a degree of any importance.

The only way in which the non-metal N will be able to maintain its potential in the case supposed here, is this that negative ions $N^{\nu'}$ from the electrolyte are deposited on the electrode, and there supply the deficit of electrons by the splitting off of electrons.

The processes that take place may be represented as follows:

$$\begin{array}{c} \uparrow \\ N_{n_P} + nr\theta_P \leftarrow nN^{\nu'}_P \\ \uparrow \\ nN^{\nu'}_L \end{array}$$

As the heterogeneous equilibrium in the boundary layer between the negative ions in the electrode and in the electrolyte must set in with very great rapidity, it is clear that it will depend on the velocity of the splitting off of electrons of the negative ions whether the electrode maintains its potential or whether it does not. Above a definite current density, i.e. above a definite velocity of withdrawal of electrons this will no longer be the case, and a consequence of this will be that the electrode will contain too few electrons and too many negative ions.

It follows from equation (7) for this case that the potential difference will become more strongly positive.

This phenomenon of polarisation is called "*supertension*" in the case of gases, but it is not essentially different from the phenomenon of anodic polarisation for metals.

It is clear that when we now proceed to the cathodic polarisation for non-metals this phenomenon should be ascribed to this that the added electrons are not quickly enough absorbed by the uncharged molecules or atoms, so the electron possesses a too high concentration of electrons, and a too small concentration of negative ions. According to our equation (7) this gives rise to a less positive value of the potential difference.

As we do not measure the potential difference, but the experi-

mental electric potential it is obvious that the polarisation phenomena should be discussed in connection with the formula that holds for the "exp. electric potential" in the case that we have to do with a non-metal. This formula is as follows:

$$E = \frac{0.058}{\nu} \log \frac{Q_N}{(N_L^{\nu'})} + 2{,}8 \quad . \quad . \quad . \quad . \quad . \quad . \quad (10)$$

in which:

$$Q = \frac{(N_L^{\nu'})}{(\theta_L)^{\nu}}.$$

From (7) and (8) follows:

$$\frac{(N_L^{\nu'})\,(\theta_P)^{\nu}}{(N_P^{\nu'})\,(\theta_L)^{\nu}} = K \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (11)$$

It has further been shown just now that with anodic polarisation (θ_P) decreases and (N_P^{ν}) increases; it follows therefore from this that in this case the *quotient of solubility* must *increase*, and the exp. electrical potential of the non-metal will therefore become more strongly positive.

For cathodic polarisation the quotient of solubility decreases, hence the experimental electrical potential of the non-metal becomes less strongly positive.

Amphibious Elements,

As has already been said the amphibious elements are characterized by this that they send both positive and negative ions into solution.

If such an element is made anode, positive ions can go into solution, or negative ions can be deposited from the electrolyte on the anode, or the two processes can take place simultaneously.

If the element is made cathode, positive ions can be deposited on the electrode, or negative ions can go into solution, or the two processes can take place side by side.

The closer study of these amphibious elements, which we hope soon to undertake, following the new theory on the electromotive equilibria, will probably give a deeper insight into the character of these so remarkable elements.

Laboratory for General and Anorganic Chemistry of the University.

Amsterdam, April 10th 1919.

Chemistry. — *"Urease and the radiation-theory of enzyme-action"*. By Dr. H. P. BARENDRECHT.

(Communicated in the meeting of May 31, 1919).

IV.

11. *Direct synthesis of urea by urease out of ammonium carbonate.*

According to the above theory the final equilibrium in the action of urease on urea will not always be found at complete hydrolysis.

In the case of a low concentration of urease the synthetic action in the outer shell will have free play.

In an alkaline urease solution, whatever its concentration, the enzyme will partially decay in the course of time, as was shown in part 9. In this case also a reverse action will manifest itself, proportional now to the concentration of urease.

To test this inference from the hypothesis, the following experiments were carried out:

In each of six large cylinders of about 1 Litre capacity 100 c.c. of water were introduced, into which 5.786 g. of ammonium carbonate were dissolved. Each of these cylinders was closed by a rubber stopper with two borings, the one carrying a straight glass tube, provided at the bottom with a bulb with pinholes, the other a bent glass tube, allowing the air-current, which was to be blown through, to pass into a second, smaller cylinder, in which the ammonia was to be absorbed. To this purpose each of these smaller cylinders contained 186,85 g. of H_2SO_4 $\frac{4}{10}$ N (which is equal to 185 c.c. $\frac{4}{10}$ N). The greater accuracy, obtained by weighing the absorbing acid, was necessary, considering that the effect looked for, was the difference of two large values and would presumably be only small.

In two of the large cylinders 3, in two others 6 g. of Soja-meal were introduced. The glass tubes of these cylinders were all closed with pieces of rubber tubing and clips. After a few hours those, making communication with the absorbing cylinders, were opened for a moment, to allow the carbonic acid, evolved by the partial change of the dissolved ammonium carbonate to ammonium carbamate, to escape through the sulphuric acid.

After leaving the six pairs of cylinders for 24 hours at room temperature, shaking through the Soja-meal from time to time, the connections with the absorbing cylinders were opened and 250 c.c. of saturated potassium carbonate was run into each of the larger cylinders through the long tubes. By then passing through for 24 hours vigorous currents of air, washed through sulphuric acid, all the ammonia was blown over into the sulphuric acid.

A few drops of octyl-alcohol, run in at the same time with the Soja-meal, prevented foaming.

The quantities of ammonium carbonate and sulphuric acid were chosen such, that only a few c.c. of NaOH $\frac{1}{10}$ N were required to titrate the free sulphuric acid, which was left.

The results of a preliminary experiment on March 14th., 1916 were as follows:

TABLE 22.

c.c. NH_3 $\frac{1}{10}$ N in:

			Mean	NH_3 converted into urea
Amm. carb. alone	735.8	735.9	735.85	
Amm. carb. + 3 g. of Soja	735.15	734.85	735.—	0.85
Amm. carb. + 6 g of Soja	734.85	734.75	734.8	1.05

Afterwards, however, it became evident, that an appreciable quantity of NH_3 was developed from the Soja-meal [1]).

In order to estimate this, in each of two large cylinders 250 c.c. of saturated potassium carbonate, 100 c.c. of water and 4 drops of octyl-alcohol were introduced. Moreover in the one 6, in the other 12 g. of Soja-meal. Each was connected with one of the small cylinders, into which 5 c.c. H_2SO_4 $\frac{2}{5}$ N and some water had been brought. After some 5 hours of blowing through the whole was left at room temperature till the next day and the blowing was started again and continued till the following day.

The 6 g. of Soja-meal had given 1,1 c.c. of NH_3 $\frac{1}{10}$ N.
The 12 g. of Soja-meal 1,65 c.c. of NH_3 $\frac{1}{10}$ N.
Hence in the mean for 6 g. of Soja-meal 0,97 c.c. $\frac{1}{10}$ N.

1) Special experiments established that no NH_3 was formed out of urea by the prolonged action of saturated potassium carbonate.

Correcting the figures of March 14th by this value we get:

ammonia converted into urea by

3 g. of Soja 1,33 c.c. $\frac{1}{10}$ N

6 g. of Soja 2.02 c.c. $\frac{1}{10}$ N.

March 28th 1916. In the large cylinders 11.572 g. of ammonium carbonate and 100 c.c. of water. In the small cylinders 190 g. of H_2SO_4 of about 0,8 N (1 gram = 7,85 c.c. $\frac{1}{10}$ N).

In two of the six large cylinders 6 g. of Soja-meal, in two others 12 grams.

After 24 hours' standing at room temperature 250 c.c. of saturated potassium carbonate and a few drops of octyl-alcohol were introduced into each and the ammonia blown over for 48 hours.

Results:

TABLE 23

c.c. NH_3 $\frac{1}{10}$ N in:

	First cylinder	Second cylinder	Mean	Corrected for NH_3 out of Soja	NH_3 converted into urea
Amm. carb. alone	1462.4	1462.5	1462.45		
Amm. carb. + 6 g. of Soja	1460.7	1460.5	1460.6	1459.6	2.85
Amm. carb. + 12 g. of Soja	1458.5	1458.4	1458.45	1456.5	5.95

May 16th 1916. A solution of about 95 g. of ammonium carbonate in 1 Litre of water was prepared and saturated with carbonic acid.

Two large cylinders, each with 100 c.c. of this solution and 250 c.c. of saturated potassium carbonate were connected with small cylinders containing each 174 g. H_2SO_4 0,8 N and all the ammonia blown over into the acid by passing an air current for 48 hours.

Four large cylinders (*A*) received each 100 c.c. of the same ammonium carbonate solution and 6 g. of Soja-meal.

Four others (*B*) each 100 c.c. of ammonium carbonate solution and 12 g. of Soja-meal.

After 2 days at room temperature one cylinder *A* and one cylinder *B* were analysed by introducing 250 c.c. of saturated potassium carbonate and blowing over the ammonia into 174 g. acid for two days. In the same way one *A* and one *B* were treated after 3 days, and two cylinders *A* and two cylinders *B* after 4 days. See Table 24.

Evidently the equilibrium is scarcely reached in about 3 days. If the activity had remained unchanged, it might have been reached much sooner, or probably no synthesis at all had been effected. According to what we have seen above, the urease had been gradually

Results:

TABLE 24.

c.c. NH_3 $\frac{1}{10}$ N in:

	First cylinder	Second cylinder	Mean	Corrected for NH_3 out of Soja	NH_3 converted into urea
Amm. carb. alone	1352.1	1352.7	1352.4		
Amm. carb. + 6 g. of Soja, 2 days	1351				
Amm. carb. + 6 g. of Soja, 3 days	1350.9				
Amm. carb. + 6 g. of Soja, 4 days	1350	1350.5	1350.3	1349.3	3.1
Amm. carb. + 12 g. of Soja, 2 days	1349.9				
Amm. carb. + 12 g. of Soja, 3 days	1348				
Amm. carb. + 12 g. of Soja, 4 days	1348.7	1348.6	1348.7	1346.7	5.7

weakened by being dissolved for such a long time in an alkaline medium and therefore a gradual displacement of the equilibrium to the side of urea was to be expected.

These experiments show clearly, that a small part of the ammonium carbonate disappears through the action of urease and that this part is proportional to the amount of urease present. Both facts are in accordance with the above theory of the synthesis of urea.

12. *The determination of H- and OH-ion concentration.*

Since the theory and results, communicated in this paper, will both have emphasized the dominating importance of frequent and therefore simple p_H determinations in enzyme research, it may be useful to give the following details of the apparatus used for this purpose.

It was nearly the same as that, described by the author as "A simple Hydrogen Electrode" in Biochem. J. 1915, 66.

The accuracy was improved by the addition of a little cup with saturated KCl solution, in which both the hydrogen-electrode and the tube of the calomel-electrode dipped. The latter was changed, as shown in Fig. 16 in order to avoid all capillaries and rubber connections. After a measurement the small flask, containing the calomel-electrode proper, was turned round and the dipping tube enclosed in a small tube with saturated KCl solution, as will be seen in the figure.

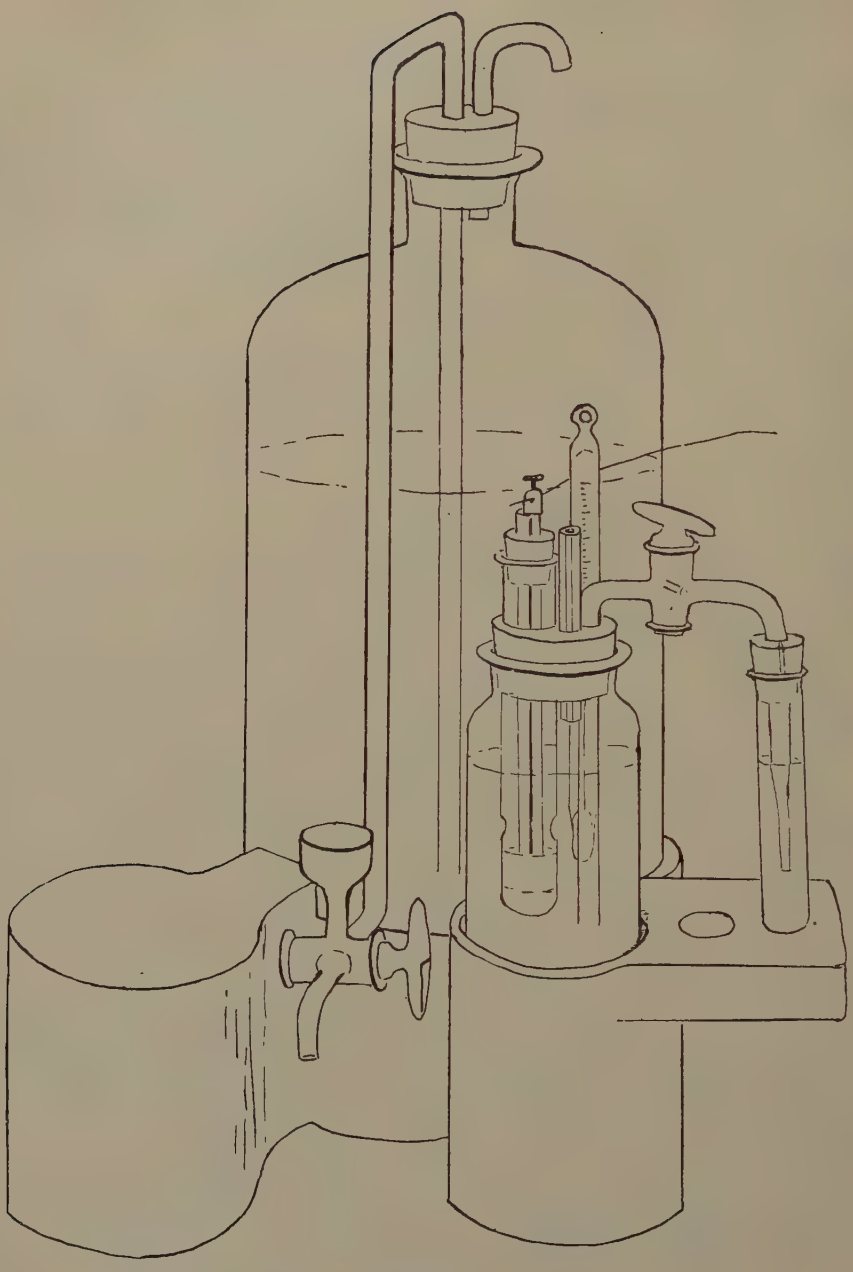

Fig. 16.

A stopcock, with a rectangular channel, allowed the small cup of about 2 c.c. capacity to be readily emptied from the large bottle with saturated KCl solution.

For the daily estimation of p_H the apparatus, represented in Fig. 17 has proved to be very satisfactory in the long run. The short perforated plunger, accurately filling up the inside of the cylinder,

when brought home, is rigidly connected to an overlapping cap, screwed on the outside of the cylinder. By turning the handle *A* backwards with cock *K* closed, cap and plunger are turned too and the latter is therefore drawn a little out of the cylinder. The liquid may thus be drawn up into the electrode tube, and by swinging the

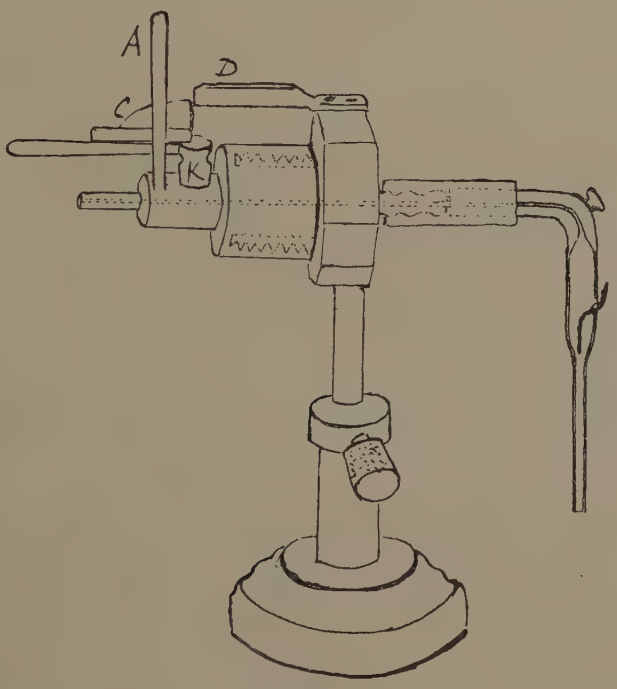

Fig. 17.

handle *A* gently to and fro the equilibrium between platinum-electrode, liquid and hydrogen can be easily established. Dead space and mistakes are avoided by the arrangements *C* and *D* on cock and frame. The protruding piece *D* allows the turning out of the plunger only, if the cock is shut and the quadrant *C* on the top of the cock is then in the position, shown in the figure. The same arrangement clearly helps to avoid the mistake of opening the cock and thus admitting the hydrogen, when the plunger is not turned home.

After shutting the cock, establishing equilibrium as described above, and adjusting the liquid in the electrode tube (by slightly turning the handle *A*) in such a way, that it just touches the point of the platinum wire, the whole apparatus is lifted up and placed on the wooden block of Fig. 16, the electrode tube dipping in the KCl-cup.

Another improvement was the carrying out of all these operations in an air-thermostat (Fig. 18), in which all the apparatus, shown

in Figures 16 and 17 as well as the hydrogen generator and wash-bottles were kept on 27°. Constancy of temperature within $\frac{1}{4}$ of a

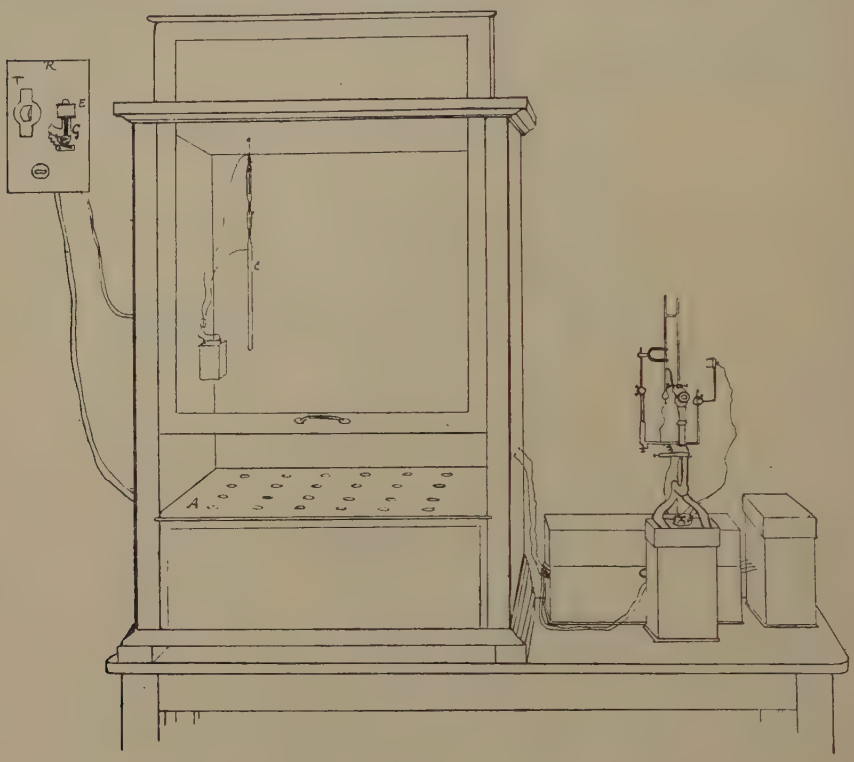

Fig. 18.

degree throughout the whole thermostat, which is amply sufficient, was attained by electrical heating in the following way.

Under the perforated bottom *A*, like the side walls and cover of the thermostat made of "eternite", a kind of concrete, some 130 metres of constantan wire of 0.4 m.m. diameter are spread out on a light frame. The current from the main of 220 Volts can be admitted to this wire by the relay *R*, an electrotechnical arrangement obtainable everywhere. A shunt of the current is reduced to some 8 Volts in the transformer *T*. This reduced current runs through the electromagnet *E* and is broken or opened by the regulator *C*. The latter is of the type, described by CLARK [1]), a spiral of nickel securing contact always at the same point of the mercury meniscus in a 2 m.m. wide capillary in pure hydrogen. To reduce sparking the poles of this regulator are also connected with a small condensor.

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[1]) J. Amer. Chem. Soc. 1913, 35, 1889.

If the current is off, the iron plunger in the evacuated glass vessel G is down and keeps the mercury in the horizontal side-tube, allowing the main current to pass between the sealed in platinum contacts. A rise of a fraction of a degree causes the large volume of mercury in the regulator C to make contact with the nickel spiral, starting the 8 volts current and thereby lifting the plunger out of the mercury in G. The running back of this mercury interrupts the main current and stops its heating effect very quickly, the heating wires being rapidly cooled, owing to their position in the open space between the table and the perforated bottom of the thermostat. Another advantage of this arrangement is the automatic stirring of the air in the thermostat by the jets of hot air rising through the holes in the bottom.

The back and front walls are double glass windows; the latter can be lifted up, allowing the necessary operations to be performed in the thermostat. These take only a few minutes. The temperature in the inside is so quickly restored after shutting the window, that the apparatus are practically maintained at the required temperature.

As will be seen in Fig. 18, the potentiometer is connected by thin wires, passing through the walls of the thermostat, with the electrode inside, with the capillary electrometer, mounted on a box, which contains an accumulator for its small lamp, and on the other side with the working accumulator and the Weston cell, contained in the third box.

As mentioned in part 5 of this paper, a direct estimation of the concentration of hydroxyl-ions, or of p_{OH}, in 8 % phosphate solutions had become indispensable.

The principle, on which these determinations were based, was the following:

By saturating a blacked platinum electrode with oxygen, an OH electrode may be obtained in the same way as a H electrode is made with hydrogen.

If π_c be the potential of the calomel electrode with saturated KCl solution and π_{OH} that of the OH electrode, the electromotive force, measured in the usual way at 27° is

$$E = \pi_c + \pi_{OH} = \pi_c + 0{,}0595 \log \frac{C}{c},$$

where C represents the concentration of OH, corresponding to the electrolytic solution pressure of the OH electrode, and c the OHion concentration. In such a cell the oxygen electrode is positive, the calomel electrode negative.

A second solution with a different hydroxyl-ion concentration gives in the same way:

$$E' = \pi_c + \pi'_{OH} = \pi_c + 0{,}0595 \log \frac{C}{c'}$$

Therefore:

$$E' - E = \pi'_{OH} - \pi_{OH} = 0{,}0595 \log \frac{c}{c'}$$

from which, substituting $p_{OH} = -\log c$ and $p'_{OH} = -\log c'$:

$$E' - E = 0{,}0595 (p'_{OH} - p_{OH}).$$

Thus, by measuring the electromotive force of the calomel electrode, combined, first with a solution of known p_{OH}, e.g. a SÖRENSEN's phosphate solution, and then with an 8 % phosphate solution, this equation for $E'—E$ gives at once the value p'_{OH}.

Determinations of this kind with two SÖRENSEN's solutions gave nearly the right value.

As is well known however, an OH electrode does not give at all as constant and accurate results as a H electrode. The potential of an OH electrode has always been found about 150 millivolts too low, which fact is commonly ascribed to the formation of some suboxyde of platinum.

This constant depression is, however, eliminated in our formula. Moreover it proved possible to arrange the experiments in such a way, that the influence of the inconstancy was repressed considerably.

Some 10 c.c. of the liquid to be examined were introduced into a small tube of about 1 cm. diameter. By dipping the electrode tube into this small quantity a quick and thorough saturation with oxygen could be obtained.

The oxygen, free from hydrogen, was washed in a bottle with 3 % KI solution to keep back possible traces of ozone. It was brought to the temperature of 27° by keeping the washbottle and rubber tubing as well as the other apparatus (the same as used for the p_H determination) in the thermostat.

After oxygen has been through for some 10 minutes, the electrode tube was lifted out of the liquid under examination and connected with the calomel electrode as usual by means of the KCl-cup; the electromotive force was read within about one minute.

This process was carried through alternately with a SÖRENSEN's solution (usually 5,6 c.c. alkaline + 4,4 c.c. acid phosphate), then with the 8 % phosphate, and then with another SÖRENSEN's solution (9 c.c. alkaline + 1 c.c. acid phosphate). Immediately after this the same series of three observations was once or twice repeated. Every estimation of the unknown p_{OH} was thus preceded and followed at

the same interval by determination in a liquid of known p^{OH}. As a gradual change in the electromotive force of the same combination was generally noticed on returning to it after the two other estimations, the influence of this change could at any rate be eliminated for the greater part by interpolation.

The value, determined in this way for the negative logarithm of k_w in 8 % phosphate solutions, was in the mean

13,78.

As to the long standing problem, why the potential of the OH electrode is usually found about 0,15 Volt too low, some information may be derived from the following observation.

If the blackened platinum electrode had been polarised kathodically in dilute sulphuric acid, the value found for its potential was too low, in accordance with the experience of previous investigators. If, however, the electrode had been the anode in sulphuric acid, the determination of its potential showed a value, by nearly the same amount too high.

The following estimations were carried out with the same 8 % phosphate solution ($p_H = 6{,}92$).

Oxygen electrode kathodically polarised:

$$\overrightarrow{\pi}_H + \overrightarrow{\pi}_{calomel} = \quad 0{,}653 \ \textit{Volt}$$

$$\overleftarrow{\pi}_{OH} + \overrightarrow{\pi}_{calomel} = -\ 0{,}421 \ \textit{Volt}$$

$$\pi_H + \pi_{OH} = \quad 1{,}08 \ \textit{Volt}$$

Oxyden electrode anodically polarised:

$$\overrightarrow{\pi}_H + \overrightarrow{\pi}_{calomel} = \quad 0{,}653 \ \textit{Volt}$$

$$\overleftarrow{\pi}_{OH} + \overrightarrow{\pi}_{calomel} = -\ 0{,}736 \ \textit{Volt}$$

$$\pi_H + \pi_{OH} = \quad 1{,}39 \ \textit{Volt}$$

The theoretical value for the electromotive force of the oxygen-hydrogen-cell at room temperature is 1,23 Volt.

This fact appears to indicate, that the difference between the potential, observed at the oxygen electrode and the theoretical value is due to a polarisation phenomenon, as it can be quantitatively reversed by reversing the state of polarisation of the electrode.

13. *General remarks.*

It will be clear from the contents of this paper, that the theoretical formula

$$\frac{nc}{0{,}434}\ log\frac{1}{1-y} + ay = mt$$

is borne out by experiment in different ways.

The hydrogen-ions having been found to absorb the enzyme radiation, the question was considered, if the hydroxyl-ions possess this property also. If such were the case, the equation should be

$$\frac{\overset{++}{n}\ c + \overset{--}{n}\ c}{0{,}434}\ log\frac{1}{1-y} + ay = mt.$$

By repeating the experiments of Part 3, but now with solutions of different p_H, in most cases $p_H = 7{,}21$, it was tried several times to decide this point. Combination of the factor $\frac{\overset{++}{n}\ c + \overset{--}{n}\ c}{0{,}434}$, found for two different p_H, should give the necessary equations to calculate both $\overset{+}{n}$ and $\overset{-}{n}$. As will be clear, however, from the considerations in Part 3, the inevitable small experimental errors have an even larger influence at a p_H below or above the $p_H = 7{,}52$ of maximum activity. It proved to be impossible to carry out experiments of sufficient accuracy for this purpose. Still, the values, obtained for $\overline{n}$, though varying widely, were generally so small, sometimes even negative, that they allowed the conclusion, that the hydroxyl-ions (i.e. negative electricity) do not absorb the urease radiation.

Since the writer's first study on Enzyme-action [1]) in 1904, in which the probability was first pointed out, that also the catalytic action of hydrogen-ions and of many other catalysts might be due to radiation, the conception of catalysis as a radiation phenomenon has been taken up from different sides.

In a series of papers LEWIS [2]) has worked out the theory, that hydrogen-ions act catalytically through radiations, a molecule of the catalysed compound becoming only reactive, if its energy has been increased by absorbed radiation (ultra red heat. radiation) to the "critical" condition.

In extensive experimental researches NILRATAN DHAR [3]) pointed

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[1]) Proc. K. Akad. Wetensch. Amsterdam and Zeitsch. physikal Chem. 49,4.

[2]) T. Chem. Soc. 1914, 2330, etc.

[3]) Proc. K. Akad. Wetensch. Amsterdam 1916 and T. Chem. Soc. 1917, 690.

out the analogy between chemical catalysts and light. His conclusion was, that: "probably the effects of increase of temperature, of light, and of chemical catalysts in a reaction are intimately connected and are possibly identical in nature". They all appeared to act by shifting the equilibrium between "active" and "inactive" molecules.

SUMMARY.

1. The enzyme urease acts by a radiation, which is only absorbed by its substrate, urea, and by H-ions.

2. The mathematical formulation of this conception is

$$-dx = m \frac{x}{x + nc} dt,$$

in which x is the concentration of urea at the time t, c the concentration and n the absorption-coëfficient of the H-ions, m a constant, proportional to the concentration of urease, if H-ion concentration as well as temperature are kept constant.

Integration gives the formula

$$\frac{nc}{0{,}434} log \frac{1}{1-y} + ay = mt,$$

in which a is the initial concentration of urea, and y the fraction of a, still present at the time t.

3. By numerous experiments this equation is shown to represent the kinetics of urease action at constant p_H and temperature. It explains the nearly straight lines of the hydrolysis in alkaline solutions equally well as the practically logarithmic curves in acid ones.

4. Comparing equal concentrations of urease at varying H-ion concentration, the constant m is found to be dependent on the p_H; i. e. the activity of a given concentration of urease is a function of the p_H of the solution.

Plotting m as a function of p_H, the resulting curve is strikingly similar to the characteristic curves for the undissociated fraction of an amphoteric electrolyte as a function of p_H.

This connection can be formulated mathematically and leads to the conclusion, that urease is an amphoteric electrolyte, whose activity is greatest when undissociated. The curve obtained represents the excess of activity of undissociated over dissociated urease.

5. This mathematical formulation leads to an approximate determination of the dissociation-constants of urease, which are calculated to be not far from those of carbonic acid and ammonia.

6. The accelerating action on urease, ascribed by previous authors

to carbonic acid, is shown to be non existent. Ammonium carbonate + carbonic acid form a powerful buffer-mixture, which can maintain constancy of p_H, the indispensable condition for constant enzyme activity in a urea solution during hydrolysis by urease.

Fresh confirmation of the above formula for the rate of hydrolysis is afforded by many experiments with this buffer-mixture.

7. The estimation of initial velocities of hydrolysis, equal concentration of urease being allowed to act on different concentrations of urea at constant p_H and T, produces results, which appear unexplainable without the radiation-theory.

The lower the p_H, the more these initial velocities increase on increase of the concentration of the urea. With high p_H there is first an increase and then a decrease on raising the urea concentration.

These facts are shown to be in perfect accordance withthe radiation-theory.

8. The influence of neutral substances is investigated experimentally and theoretically. Both decrease and increase of enzyme action by the same substance are explained by the influence the neutral body has on the dissociation-constants of water or of urease or of both of them.

9. The hypothesis is put forward that urease radiation, weakened by spreading or in any other way, causes synthesis.

Experimental evidence of this is afforded by the fact, that at high p_H, where the urease is shown to be decaying, reversion of the hydrolysis is several times observed.

10. A second inference from this conception, that outside the sphere of hydrolytic action around a urease molecule there must be a region of radiation, weakened by spreading, and therefore of synthesis, explains the fact, established by a series of new experiments, that, diluting the urease concentration beyond a certain value, its specific activity is decreased. For evidently the synthetic action of undecaying urease can only be manifested, if the spheres of hydrolytic action do not intersect each other sufficiently.

11. A third inference, that in any urease solution, in which the enzyme is decaying through the combined action of alkalinity, temperature and time, a synthesis of urea from ammonium carbonate, proportional to the urease concentration, will be observed, is tested experimentally and confirmed.

12. Description of an apparatus for the simple determination H-ion concentration at constant temperature.

The determination of the hydroxyl-ion concentration, which is needed for the calculation of the dissociation-formula of urease, is

carried out with the same apparatus, after converting the hydrogen electrode into an oxygen electrode.

Laboratory of the Dutch Yeast and Spirit Manufactory.

Delft.

A complete account of this investigation, with all experimental results, will appear in the Recueil des Travaux Chimiques des Pays-Bas.

Zoology. — "*On the phylogeny of the hair of mammals*". By Prof. P. N. VAN KAMPEN. (Communicated by Prof. WEBER).

(Communicated in the meeting of May 31, 1919).

In his work on "Die in Deutschland lebenden Arten der Saurier" (1872) and later on [1]), in a paper in which he rejects MAURER's well-known theory on the derivation of the hairs of mammals from epidermal sense-organs, LEYDIG draws the attention to the resemblance between the structure of the hairs and the so-called thigh or femoral organs of the lizards, which he considers to be a transition form between ordinary epidermal proliferations and hairs.

Less attention has been paid to this remark than it would have deserved. For the structure of the afore-mentioned organs, whose function is not known (they probably participate in the act of copulation) closely resembles in fact the structure of a hair in a simplified form [2]): they are cylindrical rods, composed of horny epidermal cells, and sunken into a follicle of the skin. They differ from hairs principally by the absence of a cutis papilla and by the fact that they do not show a differentiation in medulla, cortex and cuticle. It is true that according to MAURER [3]) they are composed of two kinds of cells, but the arrangement of these cells is quite different from the one of the elements of the hair.

As not one of the hypotheses which try to derive hairs from epidermal organs of lower Vertebrates and among which the afore-mentioned one of MAURER, based on a large body of facts, is best known, has been generally aknowledged (indeed, BOTEZAT [4]) in his review on these theories comes to the conclusion that none of them can be maintained and that the hairs in the mammals independently have taken their origin in the skin) it is desirable to examine, whether the idea uttered by LEYDIG might contain perhaps a germ of truth. Against a direct derivation of hairs from femoral organs it may be advanced that these organs among the recent reptiles

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[1]) Biol. Centralbl., XIII, 1893.

[2]) LEYDIG compares them, in my opinion wrongly, to a bundle composed of hairs glued together.

[3]) Die Epidermis und ihre Abkömmlinge. Leipzig, 1895, p. 212 ff.

[4]) Anat. Anzeiger, XLVII, 1914/15.

only occur with the Lacertilians and with these far from generally, often only with the male and, besides, always in a very limited number. Elsewhere among Reptiles, organs in some degree comparable with femoral organs are only found with crocodiles, where, according to VOELTZKOW's description [1]), they lie between the scales of the back.

But, though it would be difficult to assume the direct origin of the hairs from femoral organs, the question might still be raised, whether there might not be a connection between them, in so far that they have a common origin. If this were the case, their origin might probably be traced most easily with the last-mentioned, more simply constructed organs, which in this way might throw a light on the origin of the hair.

The morphological significance of the thighorgans has been elucidated by the research of SCHÄFER [2]). This author not only confirms what has already been recorded by earlier investigators, viz. that with Lacerta the femoral organs of the male are most strongly developed in the breeding time, but he emphatically points out that in that period no keratinisation of the cells takes place. But of more importance is what he found with Sceloporus acanthinus: with this Iguanide no horny cells are formed in the organs, but instead of them a secretion, which is composed "aus einer völlig zerfallenen, dem Secret von Talgdrüsen ähnlich sehenden Masse". SCHÄFER comes to the conclusion that the thighorgans are glandulae celluliparae, related to those sebaceous glands, which are not connected with hairs. Keratinisation occurs only, when the secretion is slow. In connection with this conclusion the statement of MAURER [3]) is of importance, that with Lacerta the contents of part of the cells of the thighorgans is of a fatty nature.

These facts point to a close connection of the femoral organs with "holocrinous" cutaneous glands, and the conclusion that they can be derived from such glands is obvious. The difference between them is not great: if the fatty secretion in a sebaceous gland were replaced by keratinisation of the cells, then a horny rod would be formed, which would show great resemblance to the thighorgans. Now the eleidin, which appears with mammals in the process of keratinisation, according to MAURER has a fatty character [4]), while on the

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[1]) Abhandl. Senckenberg. Naturf. Ges., XXVI, H. 1, 1899.

[2]) Archiv f. Naturgesch., LXVIII, Bd. I, 1902.

[3]) l. c., p. 220.

[4]) GOETTE (Arch. f. mikr. Anat., IV, 1868) also describes the occurrence of fat-globules in the young epithelial hair-germ of the sheep.

other side it is known, that the cells of the sebaceous glands of the Mammalia contain eleidin-granules and can even partly undergo keratinisation [1]). One must indeed imagine the sebaceous cells to originate from ordinary epithelium-cells, which had already the capacity of keratinisation and it is not surprising that this capacity reappears now and then.

So the femoral organs of Lacerta have arisen from cutaneous glands; they have preserved the structure of those glands, chemically however they are modified, in connection with the strong keratinisation, which is characteristic of the skin of reptiles in general.

If the femoral organs can be derived from cutaneous glands, one can imagine the same thing in the case of the hairs of mammals. Only in this case the differentiation has become greater and the structure of the organ is more complicated, in consequence of the more important function the hair has in the life of mammals. The hair papilla is to be considered of secondary origin and to have arisen in connection with the richer nutrition, which had become necessary for the stronger growth.

But there is still another phenomenon that can be easily explained by this hypothesis. The origin of the hair as a solid epidermic thickening quite agrees with that of cutaneous glands, but also with that of the femoral organs, according to the descriptions of Maurer and Schäfer. As to the thighorgans, the first author already directs the attention to this similarity with cutaneous glands of the amphibians, but attaches much importance to the difference between them, which lies in the fact that the smooth muscular fibres of the glands of the amphibians are absent in the femoral organs. In this point I cannot agree with him: these muscular fibres, which in the cutaneous glands are necessary for the extrusion of the secretion, are from their very nature superfluous in the entirely horny thighorgans, and so it is perfectly clear, that they have disappeared. And the same is true for the hairs, where they are absent as well. Another point, to which Maurer attaches much importance, is the peculiar arrangement of the matrix-cells, which appears in the very first origin of the hair and of the dermal sense organ in the same manner. It seems to me however that this arrangement may be explained by the pressure of the surrounding cells upon the growing germ and so in different cases may appear in similar circumstances.

Since the researches of De Meijere [2]) an attempt to explain the origin of the hair must take into consideration their arrangement on the skin.

[1]) Cf. Schäfer, Text-Book of Microscopic Anatomy, 1912, p. 476.
[2]) Morph. Jahrb., XXI, 1894.

In those places where in mammals scales occur, the hairs are inserted generally in groups behind them. In this point they therefore differ from the thighorgans, which are placed in the middle of scales. It is true, that not too much value ought to be attached to this fact, but yet I want to point out in the first place that the afore-mentioned dermal organs of the crocodiles, described by VOELTZKOW, are arranged between the scales, and further, that the similarity in location of hairs and femoral organs becomes greater, if the considerations of PINKUS [1]) in connection with the "hairdiscs" described by him, are right. If, as he thinks, a "Haarbezirk", that is the whole complex of scale rudiment, hair group and hairdisc, answers to the scale of reptiles, then the hairs are placed in the middle of the region of the scale, just the same as is the case with the thighorgans. PINKUS, who derives the hairdiscs from tactile spots of reptiles, cannot find an explanation for the origin of the hairs: "Das Säugetierhaar hat kein Homologen in dem Gebiet der Reptilienschuppe; sein Platz ist leer." By the hypothesis, developed before, this objection against PINKUS' theory is done away with.

MAURER has directed the attention to another arrangement of the hairs; still before the hairgroups are formed, in mammalian embryos the placing of hairs in longitudinal rows may be stated. From this fact MAURER deduces an argument for his before mentioned theory, because epidermal sense organs generally show a similar arrangement. This argument however becomes worthless by the observation

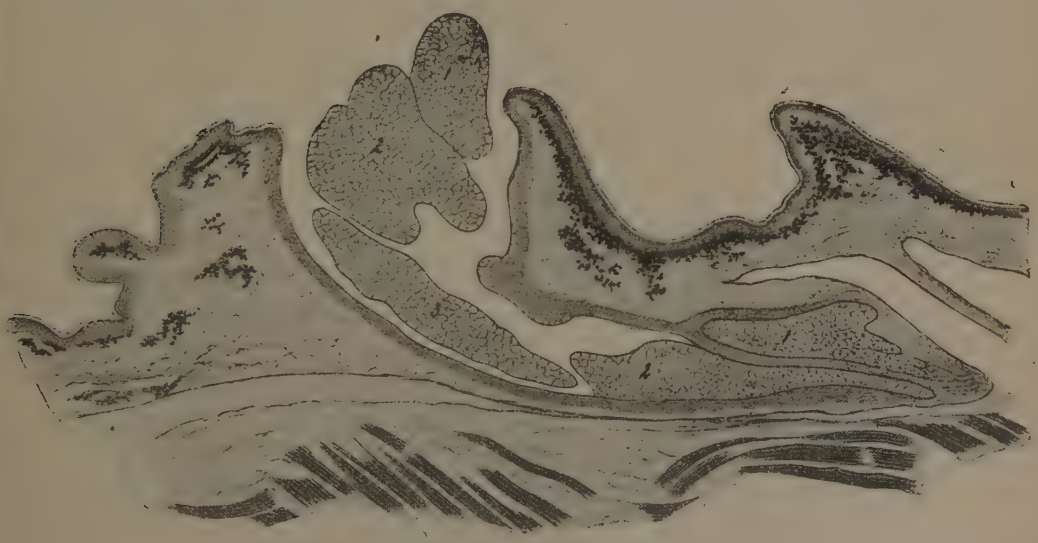

Fig. 1 Lacerta agilis. Bundle of three femoral organs (1—3).

[1]) Arch. f. mikrosk. Anat., LXV, 1905.

of MAURER himself[1]), that also the first germs of the dermal glands of Triton and Anura occur in rows.

The dermal glands of Promammalia have thus developed in the Mammalia in diverging direction: the hairs, as well as the dermal glands of the mammals have arisen from them. The complex of hair and sebaceous glands is to be derived either from a compound gland, the follicles of which have taken a different direction of development, or what seems more probable to me, from the union of a number of glands into one follicle, in the same way as hairs may be united into a bundle. This last derivation may be strengthened by the fact that a number of femoral organs too, sometimes form a bundle with a common follicle. I found this in Lacerta agilis (see fig. p. 143).

[1]) l. c., p. 159.

Physics. — *"Note on the circumstance that an electric charge moving in accordance with quantum-conditions does not radiate."* By G. NORDSTRÖM. Supplement N°. 43*a* to the Communications from the Physical Laboratory at Leiden. (Communicated by H. KAMERLINGH ONNES).

(Communicated in the meeting of May 31, 1919).

One often hears the remark: BOHR's atomic theory is at variance with classical electrodynamics in assuming that an electron which is moving according to quantum-conditions does not radiate energy in the form of electromagnetic waves. The assertion formulated in this way does not seem to me to state correctly where the opposition between BOHR's assumption and classical electrodynamics lies. In the sequel I shall try to substantiate this view. We shall begin by looking at the problem from a general point of view.

If an otherwise empty space contains electric charges whose motions are completely fixed, the electro-magnetic field is not singly determined by means of the MAXWELL-LORENTZ field-equations. In order to obtain a perfectly definite condition certain boundary-conditions must be fixed and it is to these that we shall give our attention. Whatever the field may be, it may be represented by the electro-dynamic potentials viz. the vector-potential $\mathfrak{A}$ and the scalar potential φ, which may also be combined in a four-dimensional vector-potential with the following components:

$$\varphi_x = \mathfrak{A}_x \quad , \quad \varphi_y = \mathfrak{A}_y \quad , \quad \varphi_z = \mathfrak{A}_z \quad , \quad \varphi_t = \varphi \; . \quad . \quad .) \quad 1)$$

The potentials determine the field-vectors $\mathfrak{E}$, $\mathfrak{B}$ completely by means of the equations [1])

$$\left.\begin{aligned} \mathfrak{B} &= rot\, \mathfrak{A}, \\ \mathfrak{E} &= -grad\, \varphi - \frac{1}{c}\frac{\partial \mathfrak{A}}{\partial t} \quad (c = \text{velocity of light}). \end{aligned}\right\} \quad . \quad . \quad .) \quad 2)$$

On the other hand the potentials $\mathfrak{A}$, φ are not completely determined by the field. For this reason we may submit them to the condition

$$div\, \mathfrak{A} + \frac{1}{c}\frac{\partial \varphi}{\partial t} = 0,$$

[1]) Comp. for instance M. ABRAHAM, Theorie der Elektrizität II, 2te Aufl., p. 36.

and we then obtain for the various potential-components (using rational units) the following differential equation

$$\frac{\partial^2\varphi}{\partial x^2}+\frac{\partial^2\varphi_\alpha}{\partial y^2}+\frac{\partial^2\varphi_\alpha}{dz^2}-\frac{1}{c^2}\frac{\partial^2\varphi_\alpha}{\partial t^2}=-\sigma_\alpha, \quad . \quad . \quad . \quad . \quad (3)$$

$$(\alpha = x, y, z, t).$$

In this equations σ_x, σ_y, σ_z are the components of the electric current, σ_t is the density of the electricity. The motion of the charges being given, σ_x, σ_y, σ_z, σ_t are known functions of x, y, z, t. The general solution of the differential equations (3) may then be put in the following form:

Let x_0, y_0, z_0 be the coordinates of a point for which φ_α is to be found. The distance of this point to a point (x, y, z) may be called r, so that

$$r^2=(x-x_0)^2+(y-y_0)^2+(z-z_0)^2 \quad . \quad . \quad . \quad . \quad . \quad (4)$$

The sign of r is not fixed by this relation, but we may leave it undetermined in the mean time.

A unit vector with components

$$\frac{x-x_0}{r}, \quad \frac{y-y_0}{r}, \quad \frac{z-z_0}{r} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

will be represented by r. The direction of r is evidently dependent upon the choice of the sign of r. Let F be an arbitrary closed surface which encloses the point x_0, y_0, z_0. A surface-element of F, considered as a vector directed outwards, will be denoted by $d\mathfrak{F}$. Using these symbols we may write the general integral of the differential equation (3) in the form [1]:

$$\varphi_\alpha(x_0,y_0,z_0,t)=\pm\frac{1}{4\pi}\left\{\int dV\frac{\sigma_\alpha}{r}+\int d\mathfrak{F}\left(\frac{1}{r}grad\,\varphi_\alpha+\frac{\mathrm{r}}{cr}\frac{\partial\varphi_\alpha}{\partial t}+\frac{\mathrm{r}}{r^2}\varphi_\alpha\right)\right\}_{t-\frac{r}{c}}. \quad (6)$$

The surface-integral has to be extended over the closed surface F, the space-integral over the enclosed space V containing the point x_0, y_0, z_0. The index $t-\frac{r}{c}$ is meant to indicate, that at the right-hand side the quantities σ_α, φ_α, grad φ_α, $\frac{\partial\varphi_\alpha}{\partial t}$ refer to the time $t-\frac{r}{c}$, which varies from point to point. The double sign on the right-hand

[1]) A proof of equation (6) may be found in Finska Vetenskapssocietetens Förhandl. L. I. 1908—09, Afd. A. n°. 6. If the sign of r is not fixed beforehand, it is easily found that the right-hand side has the double sign.

side is required as long as the sign of r has not been settled, but may be chosen at will; the $+$ sign holds for positive, the $-$ sign for negative r. If the sign is taken positive, the moment $t - \frac{r}{c}$ is earlier than t and φ_α must then be taken as a delayed potential. If the negative sign is chosen, the moment $t + \frac{r}{c}$ is later than t and φ_α is to be considered as an advanced potential.

Every function $\varphi_\alpha(x, y, z, t)$, therefore, which satisfies the differential equation (3), *may be considered either as a delayed or as an advanced potential, if only the contribution to the potential which is due to the boundary surface F (which may also be moved to infinity) is taken into account.*

It follows that every electro-magnetic field, i. e. every field for which the MAXWELL-LORENTZ equations hold, may be calculated for a moment t either from the condition at the time $t - \frac{(r)}{c}$ or from that at the time $t + \frac{(r)}{c}$, if only the contribution by the boundary surface is taken into account. This contribution is necessarily different in the two cases.

If the surface F is made to move to infinity and if at the same time the condition is laid down, that at the boundary the surface-integral has the value zero, if the potential is considered as a delayed potential, the ordinary solution is obtained of the problem to calculate the field from the charges. But this solution is only one particular one: there are an infinite number of others.

The author may be excused for having discussed this question at some length: it seemed to him that it is not always sufficiently kept in view.

We shall now prove, that every periodical motion of electricity allows the assumption of a field such that no energy is radiated. The separate points of the electric charges will be identified by means of 3 parameters ξ, η, ζ. Every set of values ξ, η, ζ, therefore, denotes a definite point in the electricity sharing the motion of the latter. The motion is completely described by the equations

$$\left.\begin{aligned} x &= x(\xi, \eta, \zeta, t), \\ y &= y(\xi, \eta, \zeta, t), \\ z &= z(\xi, \eta, \zeta, t), \end{aligned}\right\} \quad \ldots\ldots\ldots \quad (7)$$

that is to say, for given values of ξ, η, ζ the coordinates x, y, z are

functions of the time. Let us consider the motion represented by equations (7) as being completely given. This motion we shall call *motion* 1. We therefore assume, that x, y, z are known functions of ξ, η, ζ, t for all values of t from $-\infty$ to $+\infty$. We then calculate the electro-magnetic field by means of *delayed* potentials and choose the boundary conditions in such a manner, that the surface-integral in (6) becomes zero for each potential-component φ_x, when the surface F moves to infinity. The field is then singly determined by the motion of the electricity. We shall call the field obtained *field* 1. In this case we obviously have a radiation of energy.

We shall further consider a different motion of the electricity, *motion* 2, which is obtained from motion 1 by reversing the sign of t.

$$\left.\begin{aligned} x &= x(\xi, \eta, \zeta, -t), \\ y &= y(\xi, \eta, \zeta, -t), \\ z &= z(\xi, \eta, \zeta, -t), \end{aligned}\right\} \text{motion 2.}$$

In this system all paths are evidently described in a direction opposite to that of motion 1. For motion 2 we again calculate the electro-magnetic field by means of *delayed* potentials and with the same boundary-conditions as before. We shall again obtain a field with energy-radiation, which we shall call *field* 2.

If the motion 1 is periodical, this will also be the case for motion 2 and the radiation during one period is equal for field 1 and field 2. We now pass from field 2 to a new field 3, by reversing the sign of t and at the same time the components of the magnetic field $\mathfrak{B}_x, \mathfrak{B}_y, \mathfrak{B}_z$. It is easily shown, that with this change of sign the MAXWELL-LORENTZ field-equations remain valid. As t changes sign, the motion of the electricity is reversed. *The motion of the electricity is therefore precisely the same in field* 3 *as in field* 1. Owing to the reversal of the sign of $\mathfrak{B}_x, \mathfrak{B}_y, \mathfrak{B}_z$, ($\mathfrak{E}_x, \mathfrak{E}_y, \mathfrak{E}_z$-retaining the same sign) the direction of the energy-stream is reversed, so that in field 3 we have a radiation of energy *inwards*. For a periodical motion of the electric charges the amount of energy drawn in during a period is the same as the radiation outwards in fields 1 and 2.

It is further easily found that field 3 may be calculated from *advanced* potentials, with a zero-contribution of infinity. If on the other hand the potentials are taken as delayed, the contribution of infinity is not equal to zero.

We now superpose field 1 and 3, which is possible since the field-equations are linear. In the two fields taken separately the electricity has the same motion, which therefore remains the same in the combined field. The density of the electricity on the other

hand is everywhere doubled by the superposition. In order to reduce this to the former value we diminish the strength of the field in the combination to half its value. The density of the electricity is thereby diminished in the same ratio, and in the new *field* 4 we thus obtain precisely the same distribution and motion of the electricity as in field 1. The motion being periodical the energy-stream in field 4 evidently gives a total radiation zero during a full period. We have obtained a kind of stationary electro-magnetic waves.

Since the motion of the electricity in field 4 is identical with the motion from which we started, it has been proved that *every periodical motion of electric charges allows the assumption of an electromagnetic field without radiation of energy.* Without further enquiry it is clear, that this proposition also holds, if the motion of the charges is not exactly periodical, but consists in, say, a planetary motion with a movement of the perihelion.

The question remains, whether BOHR's theory can draw any benefit from the result arrived at, but it seems that such is not the case. If the electrons in an atom were going round in the same orbits for all eternity, there would be nothing to prevent us assuming an electro-magnetic field such as field 4. But the sudden transitions from one allowable orbit to another cause difficulties. A simple calculation shows that in field 4 the energy-density for large distances r is proportional to $\frac{1}{r^2}$, the energy of the whole field, therefore, becoming infinite. In consequence of this the change of energy associated with the transition of an electron has quite a different value to what BOHR has to assume, and it does not seem to me possible to make the two values agree. The above discussion, therefore, hardly seems to have a direct bearing on BOHR's theory, but it does seem to me to be of some use for obtaining a broader insight into the question as to where the difficulties of BOHR's theory actually lie. The result to which in my opinion it leads in this case was stated in the beginning of this note and I should like to formulate it in this way: the usual statement that it is inexplicable why an electron moving in accordance with the quantum-conditions should not radiate energy, seems to me to be based on an assumption which is not sufficiently general. A more general conception of the problem although unable to solve the difficulty, seems to me to put it in a different light.

Leiden, April 18, 1919.

Mathematics. — *"Remark on Multiple Integrals."* By Prof. L. E. J. BROUWER.

(Communicated in the meeting of June 28, 1919).

The object of this communication is to make two remarks in conjunction with the first part of my paper: *"Polydimensional Vectordistributions"* [1]) presented at the meeting of May 26, 1906.

§ 1.

I. The proof of the generalisation of STOKES's theorem given l.c. pp. 66—70 provides this generalisation not only in the Euclidean but also in the following ametric form:

THEOREM. *In the n-dimensional space* $(x_1, \ldots x_n)$ *let the* $(p-1)$*-tuple integral*

$$\int \Sigma F_{\alpha_1 \ldots \alpha_{p-1}}(x_1, \ldots x_n)\, dx_{\alpha_1} \ldots dx_{\alpha_{p-1}} \quad . \quad . \quad . \quad . \quad (1)$$

be given, where the F's are continuous and finitely and continuously differentiable; consider also the p-tuple integral

$$\int \Sigma f_{\alpha_1 \ldots \alpha_p}(x_1, \ldots x_n)\, dx_{\alpha_1} \ldots dx_{\alpha_p}, \quad . \quad . \quad . \quad . \quad . \quad (2)$$

where

$$f_{\alpha_1 \ldots \alpha_p} = \sum_{\nu=1}^{p} \frac{\partial F_{j_\nu}}{\partial x_{\alpha_\nu}}$$

$(indicatrix\ j_\nu\ \alpha_\nu\ aeq.\ indicatrix\ \alpha_1 \ldots \alpha_p)$.

Then, if the two-sided p-dimensional fragment [2]) G *is bounded by the two-sided* $(p-1)$*-dimensional closed space* g*, the indicatrices of* G *and* g *corresponding and both* G *and* g *possessing a continuously varying plane tangent space, the value of* (1) *over* g *is equal to the value of* (2) *over* G.

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[1]) See Vol. IX, pp. 66—78; we take the definitions modified in accordance with note [1]) on p. 116 l. c. I take this opportunity of pointing out that on p. 76 l.c. lines 13 and 14 "finite sourceless current system" should be read instead of "system of finite closed currents".

[2]) Math. Annalen 71, p. 306.

Of this theorem, which was enunciated by POINCARÉ [1]) in 1899 already, without proof however, and in a form expressing the rule of signs in a less simple manner, I shall here give the proof anew, editing it somewhat more precisely than in my quoted paper.

II. In the n-dimensional space $(x_1, \ldots x_n)$, which we shall denote by S, let the p-tuple integral

$$\int \Sigma\, \varphi_{\alpha_1 \ldots \alpha_p}(x_1, \ldots x_n)\, dx_{\alpha_1} \ldots dx_{\alpha_p} \quad . \quad . \quad . \quad . \quad . \quad (3)$$

be given, where the φ's are continuous.

Let Q be a two-sided p-dimensional net fragment [2]) provided with an indicatrix and situated in S, σ a base simplex of Q with the vertex indicatrix $A_1 A_2 \ldots A_p A_{p+1}$, A an arbitrary point of σ, $A\varphi_{\alpha_1 \ldots \alpha_p}$ the value of $\varphi_{\alpha_1 \ldots \alpha_p}$ at A, ${}_\nu x_\mu$ the value of x_μ at A_ν. For every σ we determine the value of

$${}_\sigma\varphi \equiv \Sigma\, A\varphi_{\alpha_1 \ldots \alpha_p} \cdot {}_\sigma i_{\alpha_1 \ldots \alpha_p};$$

where

$${}_\sigma i_{\alpha_1 \ldots \alpha_p} \equiv \frac{1}{p!} \begin{vmatrix} {}_1x_{\alpha_1} & \cdots & {}_{p+1}x_{\alpha_1} \\ \cdot & \cdots & \cdot \\ {}_1x_{\alpha_p} & \cdots & {}_{p+1}x_{\alpha_p} \\ 1 & \cdots & 1 \end{vmatrix},$$

and where, for different terms under the Σ sign A may be chosen differently; and we sum ${}_\sigma\varphi$ over the different base simplexes of Q. The upper and lower limit between which this latter sum varies on account of the free choice of the points A, we call *the upper and lower value of* (3) *over* Q.

If we now subject Q to a sequence of indefinitely condensing simplicial divisions which give rise to a sequence Q', Q'', ... of net fragments covering Q, then, as ν increases indefinitely, the upper and lower value of (3) over $Q^{(\nu)}$ converge to the same limit, which we call *the value of* (3) *over* Q.

Let F be a two-sided p-dimensional fragment provided with an

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[1]) Les méthodes nouvelles de la mécanique céleste III, p. 10. The significance of the rule of signs here formulated, is apparent only after comparing former publications of the same author from the Acta Mathematica and the Journal de l'École Polytechnique in which the equivalence of the identically vanishing of (2) and the vanishing of (1) over every g was pronounced.

[2]) Math. Annalen 71, p. 316.

indicatrix and situated in S, f a sequence of indefinitely condensing simplicial approximations P', P'', ... of F corresponding to a category ψ of simplicial divisions. If the values of (3) over P', P'', ... converge to a limit which is independent of the choice of f so far as it is left free by ψ, then we call this limit *the value of* (3) *over* F *for* ψ.

III. We shall now occupy ourselves with the value of (1) over the boundary β of a p-dimensional simplex σ provided with an indicatrix and situated in S. In doing so we take it that the indicatrices of β and σ *correspond*, that is to say, the indicatrix of an arbitrary $(p-1)$-dimensional side of σ is obtained by placing the vertex of σ which does not belong to this side last in the indicatrix of σ and subsequently omitting it. We begin by confining ourselves to the contribution of the single term

$$\int F_{\alpha_1 \ldots \alpha_{p-1}} \, dx_{\alpha_1} \ldots dx_{\alpha_{p-1}}$$

to the value of (1) over β. By a suitable simplicial division ζ of the space $(x_{\alpha_1}, \ldots x_{\alpha_{p-1}})$ we determine a simplicial division of β, whose base simplexes correspond in pairs to those of $(x_{\alpha_1}, \ldots x_{\alpha_{p-1}})$. The family of those $(n-p+1)$-dimensional spaces within which $x_{\alpha_1}, \ldots x_{\alpha_{p-1}}$ are constant, cuts the plane p-dimensional space in which σ is contained, in a family of straight lines which connect pairs of corresponding base simplexes of β into p-dimensional truncated simplicial prisms. If e_1 and e_2 are a pair of corresponding base simplexes of β, d the concomitant truncated simplicial prism, l a line segment having components $r_{\alpha_p}, \ldots r_{\alpha_n}$ which leads from a point of e_1 to the corresponding point of e_2, then the contribution of the term $\int F_{\alpha_1 \ldots \alpha_{p-1}} \, dx_{\alpha_1} \ldots dx_{\alpha_{p-1}}$ to the value of (1) over e_1 and e_2 becomes

$$- {}_{e_1}i_{\alpha_1 \ldots \alpha_{p-1}} \cdot \sum_{\nu=p}^{n} r_{\alpha_\nu} \cdot A \left\{ \frac{\partial F_{\alpha_1 \ldots \alpha_{p-1}}}{\partial x_{\alpha_\nu}} \right\} + \varepsilon,$$

where A denotes a point of σ which may be different for the different terms under the Σ sign, and ε becomes indefinitely small with respect to ${}_{e_1}i_{\alpha_1 \ldots \alpha_{p-1}}$ for indefinite condensation of ζ.

Now let $B_1 B_2 \ldots B_p$ be a vertex indicatrix of e_1 and ${}_\nu x_\mu$ the value of x_μ at B_ν, then the value of $r_{\alpha_\nu} \cdot {}_{e_1}i_{\alpha_1 \ldots \alpha_{p-1}}$ can be expressed as

$$\frac{1}{(p-1)!}\begin{vmatrix} {}_1x_{\alpha_1}-{}_px_{\alpha_1} & \cdots & {}_{p-1}x_{\alpha_1}-{}_px_{\alpha_1} & 0 \\ \cdots & \cdots & \cdots & \cdots \\ {}_1x_{\alpha_{p-1}}-{}_px_{\alpha_{p-1}} & \cdots & {}_{p-1}x_{\alpha_{p-1}}-{}_px_{\alpha_{p-1}} & 0 \\ {}_1x_{\alpha_\nu}-{}_px_{\alpha_\nu} & \cdots & {}_{p-1}x_{\alpha_\nu}-{}_px_{\alpha_\nu} & r_{\alpha_\nu} \end{vmatrix},$$

thus also as

$$-d^{i}_{\alpha_1\ldots\alpha_{p-1}\alpha_\nu}+\varepsilon$$

so that the contribution of the term $\int F_{\alpha_1\ldots\alpha_{p-1}}\,dx_{\alpha_1}\ldots dx_{\alpha_{p-1}}$ to the value of (1) over e_1 and e_2 can be expressed as

$$\varepsilon+\sum_{\nu=p}^{n}{}_A\left\{\frac{\partial F_{\alpha_1\ldots\alpha_{p-1}}}{\partial x_{\alpha_\nu}}\right\}d^{i}_{\alpha_1\ldots\alpha_{p-1}\alpha_\nu},$$

and the value of (1) over β is obtained in the form:

$$\sum_{\alpha_1\ldots\alpha_p}\sum_{\nu=1}^{p}{}_A\left\{\frac{\partial F_{j_\nu}}{\partial x_{\alpha_\nu}}\right\}\sigma^{i\alpha_1\ldots\alpha_p}$$

(*indicatrix* $j_\nu\,\alpha_\nu$ *aeq. indicatrix* $\alpha_1\ldots\alpha_p$),

where A represents a point of σ which may be different for different terms under the $\Sigma\Sigma$ sign.

Hence it follows immediately, that, if Q is a two-sided p-dimensional net fragment situated in S, and R denotes the boundary of Q, while the indicatrices of Q and R correspond, then *the value of* (1) *over* R *is equal to the value of* (2) *over* Q.

IV. To complete the proof of the theorem formulated in I, we consider a category ψ of simplicial divisions of g such that the aggregate of the base sides possesses for ψ uniformly continuously varying plane tangent spaces, and the ratio of the volume of a base simplex to the $(p-1)^{\text{th}}$ power of its greatest coordinate fluctuation does not fall below a certain minimum for ψ. Let $g', g'', \ldots$ be a sequence of indefinitely condensing simplicial approximations of g corresponding to ψ. If, on $g^{(\mu)}$ we construct an approximating simplicial representation $g^{(\nu\mu)}$ of $g^{(\nu)}$, then, by choosing both μ and ν above a suitable limit, we can, in virtue of III, see to it that the values of (1) over $g^{(\nu\mu)}$ and $g^{(\nu)}$ differ from each other by as little as we please, whilst $g^{(\mu)}$ is covered by $g^{(\nu\mu)}$ with degree one, so that the values of (1) over $g^{(\mu)}$ and $g^{(\nu\mu)}$ are equal. Thus there exists a *value of* (1) *over* g *for* ψ which, naturally, does not change if, instead of ψ, some other category of the same kind is chosen.

Now let χ be a category of simplicial divisions of G analogous to ψ. The resulting simplicial divisions of g belong to a category ψ of the kind just described. Let $G', G'', \ldots$ be a sequence of indefinitely condensing simplicial approximations of G corresponding to χ, then, at the same time, there is hereby determined a sequence $g', g'', \ldots$ of indefinitely condensing simplicial approximations of g corresponding to ψ. Since, in virtue of III, the value of (1) over $g^{(\nu)}$ is equal to the value of (2) over $G^{(\nu)}$, *there exists, just as there does a value of* (1) *over* g *for* ψ, *a value of* (2) *over* G *for* χ, *both values being equal, and not changing if some other category of the same kind is chosen in the place of either* ψ *or* χ.

§ 2.

In introducing l.c. p. 70 the notion of a second derivative, we have omitted to give the definition of the underlying concept of *normality* of an S_p provided with an indicatrix and an S_{n-p} provided with an indicatrix which are perpendicular to each other in an S_n provided with an indicatrix. This definition we shall here give.

Let T be the point of intersection of S_p and S_{n-p}, $\alpha_1 \ldots \alpha_p T$ the indicatrix of S_p and $\beta_1 \ldots \beta_{n-p} T$ the indicatrix of S_{n-p}; we call S_p *normal to* S_{n-p} and S_{n-p} *postnormal to* S_p, if $\alpha_1 \ldots \alpha_p T \beta_{n-p} \ldots \beta_1$ is an indicatrix of S_n.

Thus, for some values of n the concepts *normal* and *postnormal* are equivalent, for other values not equivalent.

Furthermore we call a p-dimensional vector system V *normal* to an $(n-p)$-dimensional vector system W at the same point, and W *postnormal* to V, if, with respect to a rectangular system of coordinates the components of V are respectively normal to and of equal scalar values as the components of W.

In this terminology, the second derivative of the vector distribution X is the normal distribution of the first derivative of the postnormal distribution of $'X$.